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FINAL REPORT

DEVELOPMENT OF ADVANCED HIGH STRENGTH TANTALUM BASE ALLOYS PART II - SCALE-UP INVESTIGATION

BY

R.L.AMMON AND R.W.BUCKMAN.JR.

PREPARED FOR

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

CONTRACT NAS 3-10939

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FOREWORD

The work described in this report was performed by the Westinghouse Electric Corporation, Astronuclear Division. Technical administration at the Astronuclear Laboratory was under the direction of Messrs. R. T. Begley and R. W. Buckman, Jr., while Mr. P. Moorhead served as the NASA Project Manager. The period covered by the work described was from October 1967 through July 4, 1970.

ABSTRACT

Three experimental tantalum alloy compositions containing 14-16% W, 1% Re, 0.7% Hf, 0.025% C or 0.015% C and 0.015% N were prepared as two inch diameter ingot by consumable electrode vacuum arc melting. The as-cast ingots were processed by extrusion and swaging to one inch and 0.4 inch diameter rod and evaluated. Excellent high temperature forging behavior was exhibited by all three compositions. Creep strength at 2000°F to 2400°F was enhanced by higher tungsten additions as well as substitution of nitrogen for carbon. Weldability of all three compositions was determined to be adequate. Room temperature ductility was retained in the advanced tantalum alloy compositions as well as a notched/unnotched strength ratio of 1.4 for a notched bar having a $K_t = 2.9$.

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1.0 SUMMARY OF RESULTS

Three experimental tantalum alloy compositions were prepared as two inch diameter ingot and had the following compositions (Heat Number):

Ta-14W-1Re-0.7Hf-0.025C (NASVF-100)
Ta-16W-1Re-0.7Hf-0.025C (NASVF-200)
Ta-14W-1.5Re-0.7Hf-0.015C-0.015N (NASVF-300)

The basis for selecting these compositions was covered in detail in NASA CR-120818 which was the screening phase investigation culminating in this scale-up investigation. All three compositions were processed to one inch and 0.4 inch diameter rod by a combination of extrusion and swaging at 2550°F.

Both carbide strengthened alloys exhibited tensile and creep properties superior to ASTAR-811C (Ta-8W-1Re-0.7Hf-0.025C) in almost direct relation to the tungsten content. For example, at 2200°F ASTAR-811C will creep 1% in 1000 hours at a stress of 17,500 psi while NASVF-100 and NASVF-200 require 26,000 psi and 32,000 psi respectively. NASVF-300 exhibited creep properties similar to NASVF-200 at 2000-2400°F indicating that substitution of one half of the carbon with nitrogen is as effective as two atom percent tungsten on high temperature creep strength. Forging reductions of 2:1 and 3:1 were sustained by all three compositions at 2200, 2500, and 2800°F without failure.

Limited electron beam welding evaluation indicated all three compositions could be welded satisfactorily. Behavior of NASVF-200 and -300 however showed that post weld annealing would be required for more reliable properties.

2.0 INTRODUCTION

The primary objective of the work described in this report was the development of tantalum base alloy(s) exhibiting higher mechanical strength than ASTAR-811C[1]. ASTAR-811C (Ta-8W-1Re-1Hf-0.025C), developed under contract NAS 3-2542, is a fabricable, weldable sheet alloy which has significantly better creep resistance than any of the commercially available tantalum alloys such as T-111 and Ta-10W[1,2,3]. The level of strengthening additions to the ASTAR-811C composition was limited by fabricability and weldability considerations. However, it was apparent during this prior investigation that relaxation of the weldability criterion could result in higher elevated temperature strength alloys which would be competitive with the high strength columbium modified TZM molybdenum base alloy. It was with this purpose that development of high strength tantalum base alloys was continued under contract NAS 3-10939. The alloy development was conducted in two sequential phases. During the initial phase, five compositions were selected and prepared as 1.4 inch diameter consumable electrode melted ingots which were processed to 3/8 inch diameter rod and evaluated. Three compositions were selected for scale-up to two inch diameter ingot based on the results of the Phase I screening investigation. The three compositions* and their assigned heat numbers** are:

Ta-14W-1Re-0.7Hf-0.025C (NASVF-100)
Ta-16W-1Re-0.7Hf-0.025C (NASVF-200)
Ta-14W-1.5Re-0.7Hf-0.015C-0.015N (NASVF-300)

The two carbide strengthened compositions NASVF-100 and NASVF-200 differ by two percent tungsten and therefore represent a compositional range for an advanced tantalum alloy with a combination of high temperature strength and low temperature ductility. The third composition, NASVF-300 has both carbon and nitrogen and the dispersed second phase is

* All compositions given in weight percent although values for W, Re, and Hf are also essentially same values in atom percent.

** The heat number will be used throughout the report discussion in referring to these compositions.

a hafnium carbonitride. The kinetics of the carbonitride precipitation reaction has been shown to enhance high temperature creep strength[1]. Detailed discussion of the investigative results leading to the selection of the three scale up compositions is in reference 4.

3.0 GENERAL EXPERIMENTAL PROCEDURES

3.1 Alloy Consolidation

The experimental alloy compositions were prepared as 2.0 inch diameter by 4 inch long ingots using non-consumable and consumable electrode melting techniques. Electrodes for consumable vacuum arc melting were prepared by a non-consumable trough melting technique. Ten 200 gram charges of accurately weighed alloy constituents were equally spaced along the length of the water cooled copper trough. The melting chamber was vacuum purged to $< 1 \times 10^{-5}$ torr, leak checked, and then backfilled with a partial pressure of high purity argon. The charges were then non-consumably melted using DC power. The resulting bar was turned over and remelted. This remelting procedure was repeated until each side had been melted three times to increase complete homogenization of the alloy constituents. The resulting bars were one inch wide by 22 inches long and approximately 0.3 inch thick. Two such bars tack welded together were used as a second melt electrode for one ingot. The second melt electrodes were consumably arc melted into a 2 inch diameter mold using AC power.

3.2 Primary and Secondary Working

Each ingot was processed to 0.4 inch diameter bar by a combination of extrusion and swaging. The top and bottom of the as cast ingots were trimmed and the side walls were conditioned to produce 1.780 inch diameter ingots which ranged in length from 3.5 to 4.5 inches. The top of the billets were machined to give a 1/4 inch bevel at 45 degrees to serve as a nose for extrusion. The billets were then plasma coated with a 0.020 inch thick layer of molybdenum. The molybdenum coating provided protection against oxidation during transfer of billet from the induction heater to the extrusion press. Billets were heated by induction under a flowing argon cover gas to 2550°F. After soaking ten minutes, the heated billet was transferred to the container of a model 1220C Dynapak (HERF) and extruded to round bar through a zirconia coated die with a 0.940 inch diameter opening.

The extrusions were trimmed to remove the nose and tail sections then chemically cleaned to remove the remaining molybdenum clad. The extrusions were recrystallized by heat treating one hour at 3000°F in a vacuum of 1×10^{-5} torr. The annealed bars were machined to a maximum defect free OD. The bars were plasma sprayed with molybdenum to a thickness of 0.020 inch for protection during the swaging operation. The molybdenum clad bars were first heated to 1800°F in an argon purged retort then transferred to a hydrogen atmosphere furnace at 2550°F. At this temperature the solubility of hydrogen in tantalum is < 30 ppm. The bars were swaged out of the hydrogen furnace to a final diameter of 0.4 inch. Reductions per pass were on the order 10 to 12 per cent initially with final reductions on the order of 15 to 18 per cent per pass.

3.3 Mechanical Property Testing

Shoulder loaded round bar test specimens with a 0.1 inch uniform diameter gage length of one inch were used for the mechanical property evaluations. Short time tensile properties were determined at a constant strain rate of 0.05 in/minute. Elevated temperature tensile testing was done at $\leq 1 \times 10^{-5}$ torr in a self resistance heated split tungsten element cold wall vacuum furnace. All creep testing was done at $< 1 \times 10^{-8}$ torr in sputter ion pumped units of the type described by Buckman and Hetherington.[5]

3.4 Heat Treatment

All heat treatments were performed in cold wall tantalum resistance heated vacuum furnaces at pressures of $\leq 1 \times 10^{-5}$ torr. Prior to annealing all specimens were pickled in a solution of equal parts of H_2O-HF-HNO_3 to ensure removal of any contaminated layers. All annealing specimens were then wrapped with 0.002 inch thick chemically cleaned tantalum foil to further minimize any possibility of contamination.

4.0 EXPERIMENTAL RESULTS AND DISCUSSIONS

4.1 Melting

Two heats of each alloy composition were prepared and melted as two inch diameter ingots. Electrodes for vacuum arc melting were fabricated by the non-consumable technique described in detail in the previous section. Vendor analysis of the starting material are given in Table 1. Two bars produced by the trough-melting technique were tack welded together to form the second melt electrode for one ingot. The second melt electrodes were consumably arc melted into a two inch diameter water cooled copper mold using AC power. Melting parameters for the consumable melts are given in Table 2. Chemical analysis of the cast ingots are given in Table 3. Target levels for alloying additions were achieved for all ingots. Interstitial levels which are by nature difficult to control were held within tolerable limits.

4.2 As-Cast Microstructures

The as-cast microstructures of the Ta-14W-1.0Re-0.7Hf-0.025C (NASVF-100) and the Ta-16W-1.0Re-0.7Hf-0.0250C (NASVF-200) alloys are typical of the ASTAR alloy series. The tantalum, tungsten, rhenium and hafnium constituents form a single phase matrix with a dispersed second phase of Ta_2C.[1] A typical "as-cast" microstructure is shown in Figure 1a. The "as-cast" microstructure of the Ta-14W-1.5Re-0.7Hf-0.015C-0.015N (NASVF-300) alloy is shown in Figure 1b. At the lower magnification, the microstructure appears single phase, however at the higher magnification, traces of a second phase can be seen. This phase is most likely Ta_2C which has been identified in similar alloys containing intentional additions of both carbon and nitrogen. Formation of the FCC carbonitride will occur during elevated temperature exposure.

4.3 Primary and Secondary Working

The cast ingots were machined to a 1.780 inch diameter in preparation for extrusion. The billet nose was given a 1/4 inch bevel at a 45 degree angle. The billets were plasma coated with a 0.020 inch thick layer of molybdenum. The billets were extruded to solid round bar

Table 1. Vendor Analysis of Starting Material

Material	Supplier	Form	Analysis, ppm											
			C	O	N	Cb	Fe	Si	Mo	Zr	Ti	Hf	Re	W
Tantalum	Wah Chang	Plare	<30	<50	26	412	<15	<20	<10	70	11	--	--	385
Ta-10W	Fansteel	Plate	10	10	10	<500	40	<10	150	--	<10	--	--	9.95%
Hafnium	Carborundum	Plate	80	100	16	---	40	--	--	2.1%	--	--	--	---
Tungsten	Fansteel	Strip	--		--	---	--	--	--	--	--	--	--	99.95%*
Rhenium	Chase-Brass	Strip	--		--	---	--	--	--	--	--	--	99.99%*	--
Tantalum Carbide	Kenametal	-100, +200 mesh powder	6.12%		--	---	--	--	--	--	--	--	--	--

* Specification minimum

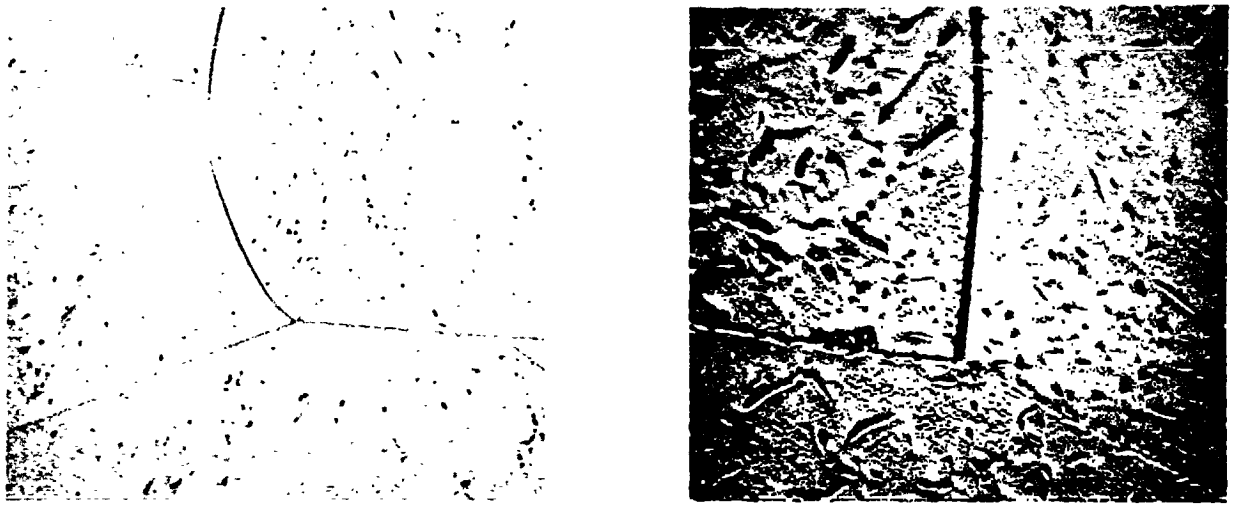
Table 2. Melt Data for Advanced Tantalum Alloys

Ingot Identity	Melt Parameters		
	AC Volts	Amps	Melt Rate (lbs/min)
NASVF-100A	25	1800	2.4
NASVF-100B	28	1350	2.3
NASVF-200A[1]	30	1800	--
NASVF-200B	30	1800	2.4
NASVF-300A	30	1800	2.1
NASVF-300B[2]	25	1950	

1. Melt was interrupted - electrode moved too near mold - melt restarted under partial pressure of argon and concluded under vacuum.
2. Melted under partial pressure of argon.

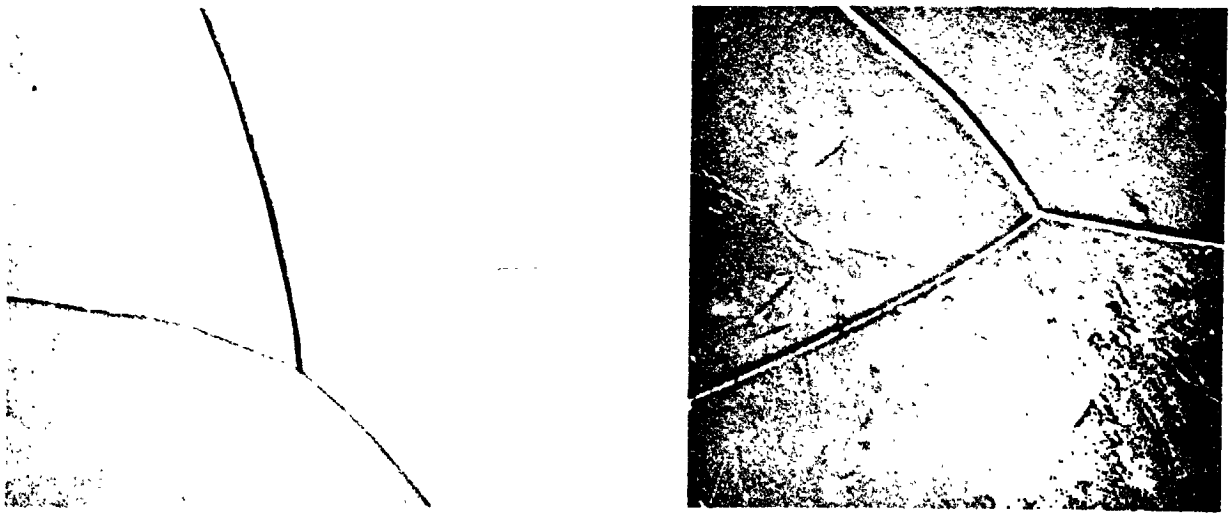
Table 3. Chemical Analysis of Advanced Tantalum Alloys

Ingot Identification and Sample Location	Analysis, Weight Per Cent					
	W	Re	Hf	C	O	N
Ta-14W-1.0Re-0.7Hf-0.025C						
NASVF-100A Top	13.5	1.10	0.69	0.0270	--	0.0016
NASVF-100A Bot	13.0	1.20	0.80	0.0300	0.0020	0.0017
NASVF-100B Top	13.5	1.00	0.65	0.0240	--	0.0050
NASVF-100B Bot	13.6	1.00	0.68	0.0240	--	0.0050
Ta-16W-1.0Re-0.7Hf-0.025C						
NASVF-200A Top	15.5	0.90	0.65	0.0180	0.0025	0.0014
NASVF-200B Bot	15.6	1.05	0.70	0.0270	0.0017	0.0010
Ta-14W-1.5Re-0.7Hf-0.015C-0.015N						
NASVF-300A Bot	13.6	1.43	0.77	0.0170	0.0032	0.0105
NASVF-300B Top	14.1	1.52	0.54	0.0140	0.0018	0.0158
NASVF-300B Bot	14.5	1.50	0.87	--	--	--



500X 1500X

a) NASVF-100



500X 1500X

b) NASVF-300

Figure 1. As Cast Microstructure of NASVF-100 and NASVF-300

approximately one inch in diameter. A Dynapak, a high-energy-rate-machine, was used to perform the extrusion operation. Extrusion data are given in the following Table 4.

Table 4. Extrusion Data for Advanced Tantalum Alloys

Billet Identification	Billet Size[1] (Length-Inches)	Extrusion Temperature (°F)	Extruded Size	Extrusion Ratio[2]
NASVF-100A	4.5	2550	1.04 x 13-3/4	2.9:1
NASVF-100B	4.5	2550	0.99 x 14-1/2	3.1:1
NASVF-200A	4.0	2550	1.07 x 11-1/2	2.8:1
NASVF-200B	4.0	2550	1.12 x 10-1/2	2.5:1
NASVF-300A	4.5	2550	1.02 x 13-3/4	3.0:1
NASVF-300B	3.6	2550	1.04 x 11	2.9:1

1 Diameter of all conditioned billets 1.780 inches
Billets plasma spray coated with 0.020 inch molybdenum coating

2 Reduction ratio = $\frac{\text{area of Billet}}{\text{area of Extrusion}}$

The co-extruded molybdenum clad was chemically removed from the extruded bars. The bars were then centerless ground to a uniform diameter. The bars were given a recrystallization anneal of one hour at 3000°F. Approximately, 7 inches of extruded bar of each alloy composition were set aside for the forging evaluation. The remainder of the extruded bar stock was plasma spray coated with molybdenum in preparation for swaging to 3/8 inch diameter rod. The swaging was accomplished at 2550°F out of a hydrogen atmosphere furnace. The bars were preheated in an argon purged furnace to a temperature of 1600 to 1700°F and then transferred hot to the hydrogen furnace which was operating at the swaging temperature of 2550°F. Reductions on the order of 10 to 15 per cent were taken per swaging pass. The rods were returned to the hydrogen furnace between swaging passes. During the swaging of NASVF-100A and B and NASVF-200A and B material, the molybdenum coating spalled off leaving the rods unprotected. Swaging was halted at a diameter of approximately 0.8 inches.

The rods were conditioned by centerless grinding, annealed one hour at 3000°F, and recoated with plasma sprayed molybdenum. Swaging was continued at 2550°F using the previously described procedure. Approximately 30 inches of 0.400 inch diameter rod was produced of each alloy composition, NASVF-100 and 200. Alloy NASVF-300, Ta-14W-1.5Re-0.7Hf-0.015C-0.015N, could not be swaged under the conditions described. One piece, NASVF-300A, broke into many fragments at a diameter of 0.6 inch. The second piece, NASVF-300B fractured on the first swaging pass and exhibited many additional cracks along the bar length. Only one piece of swaged material with sufficient length to make a creep specimen was salvaged.

4.4 Recrystallization Behavior

The alloys, NASVF-100 and NASVF-200, which were successfully swaged to 0.4 inch diameter rod at 2550°F were evaluated to determine recrystallization behavior. Samples of NASVF-300 which broke up during swaging were also included in this study. Room temperature hardness measurements and optical metallography were used to assess the response of the swaged alloy rod to thermal treatment. The samples were thermally exposed for one hour at temperatures in the range 2000 to 3600°F. Hardness values are given in Table 5 and are shown graphically in Figure 2.

The hardness behavior of the swaged rod in response to heat treatments is influenced by working temperature and the amount of reduction. The alloys were swaged at 2550°F, slightly below the recrystallization temperature. Alloys, NASVF-100 and 200 were reduced approximately 74 per cent after an intermediate recrystallization anneal to a finish diameter of 0.4 inch. NASVF-300 was reduced approximately 60 per cent prior to the break up of the material. The as-swaged hardness values reflect the recovery which occurred during reheat between swage passes and during the one-half hour anneal at the swaging temperature upon completion of swaging. Alloys NASVF-100 and 200 exhibited hardness values of 405 and 416 DPH respectively in the as swaged condition indicating a fair amount of retained strain energy even though the material was worked at 2550°F. Both alloys show little effect

Table 5. Effect of Annealing Temperature on the Room Temperature Hardness of Swaged Tantalum Alloy Rods

Condition	NASVF-100	NASVF-200	NASVF-300
As Swaged	405(1)	416(1)	442(2)
Swaged + 1 hr/2192°F	342	---	---
Swaged + 1 hr/2552°F	349	354	374
Swaged + 1 hr/2732°F	---	354	382
Swaged + 1 hr/3000°F	351	361	397
Swaged + 1 hr/3272°F	347	362	395
Swaged + 1 hr/3632°F	352	370	285

(1) 74% reduction at 2550°F

(2) 60% reduction at 2550°F

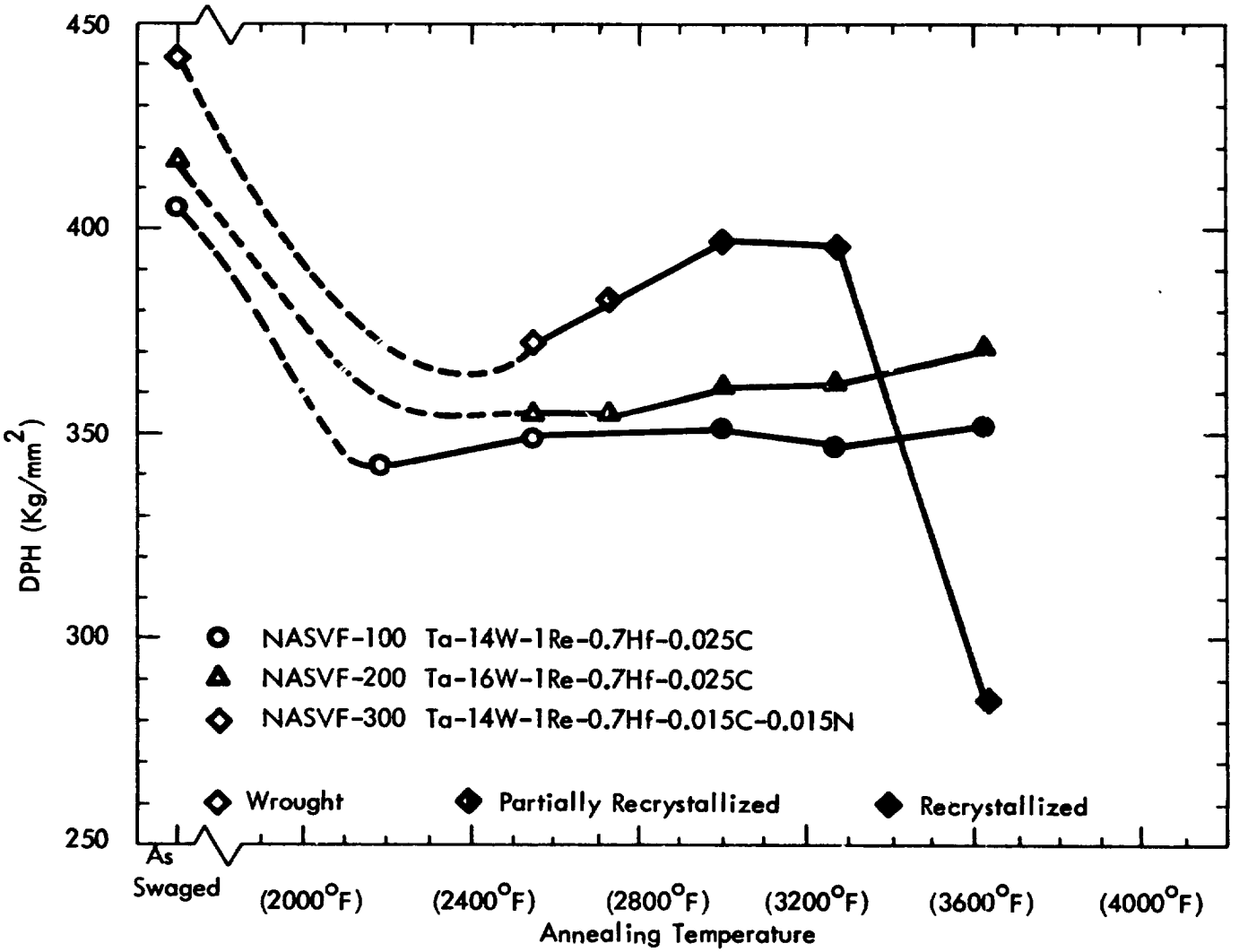


FIGURE 2 - Room Temperature Hardness of Swaged Rod as a Function of One Hour Anneals at Various Temperatures

of annealing treatments aside from the initial recovery of strain energy. NASVF-100 hardness values varied between 342 DPH after one hour at 2192°F to 352 DPH after one hour at 3632°F. NASVF-200 which contains 2 per cent higher tungsten content displayed hardness values comparable to NASVF-100 after one hour at 2552°F. Successive anneals at higher temperatures produced hardness values for NASVF-200 which were 10 to 20 DPH higher than NASVF-100. The hardness behavior of both alloys in response to heat treatments was not typical of tantalum alloys of this type.

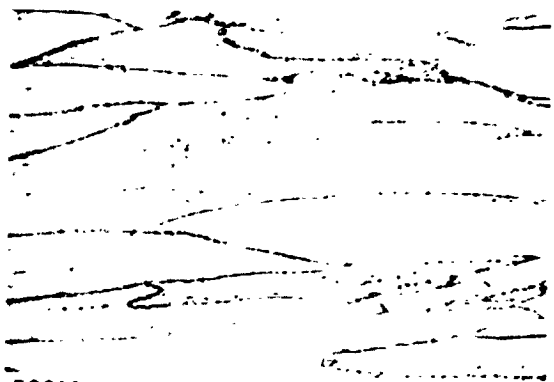
Prior work has shown that a significant reduction in room temperature hardness occurs after annealing at temperatures below the recrystallization temperature. The hardness increases with annealing temperatures up to 3000°F then remains constant to 3300°F, with a possible slight increase at 3600°F depending on the cooling rate from the annealing temperature. The abrupt increase in hardness begins at the temperature where recrystallization is initiated for one hour at temperature. After one hour at 3000°F, recrystallization is completed and a grain size of ASTM 6-8 is attained. Alloys NASVF-100 and NASVF-200 exhibited similar microstructural behavior with recrystallization complete after one hour at 3000°F.

The hardness behavior of NASVF-300 was typical for a tantalum alloy containing nitrogen and carbon as intentional alloy additions. The as swaged hardness was higher than the other two alloys although the total reduction, approximately 60 per cent, was lower. The hardness increased with increasing annealing temperature except at 3632°F, where significant reduction in hardness occurred. The drastic reduction in hardness is attributed to an overageing reaction or to the loss of nitrogen during vacuum annealing. Most likely a combination of both factors is responsible for the hardness behavior.

The recrystallization behavior of the three alloys is illustrated by the microstructures shown in Figures 3, 4, and 5. Tantalum alloys heavily worked at room temperature or slightly above, generally recrystallizes in the temperature range 2400 to 2600°F. As can be seen in

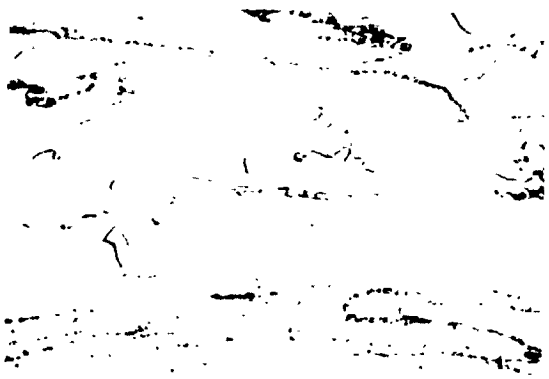
500X DPH 405

A. As Swaged



500X DPH 342

B. Swaged + 1 Hr/ 2552°F



500X DPH 349

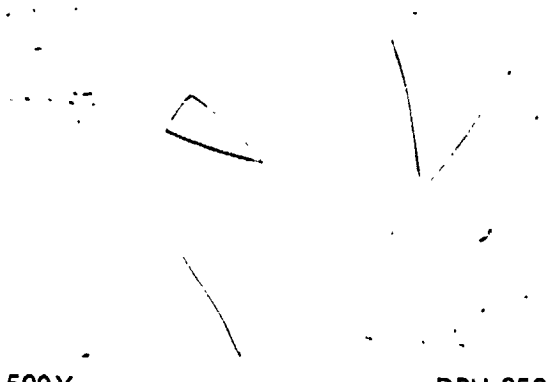
C. Swaged + 1 Hr/ 2732°F

500X DPH 351

D. Swaged + 1 Hr/ 3000°F

500X DPH 347

E. Swaged + 1 Hr/ 3272°F



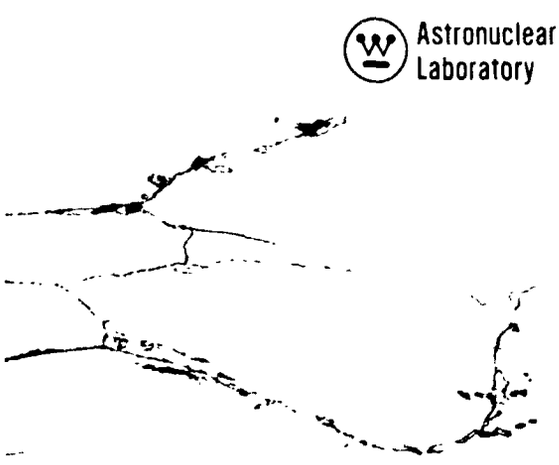
500X DPH 352

F. Swaged + 1 Hr/ 3632°F

Figure 3. Microstructures of NASVF-100 (Ta-14W-1Re-0.7Hf-0.025C) Swaged and Heat Treated at Various Temperatures

500X DPH 416

A. As Swaged



500X DPH 354

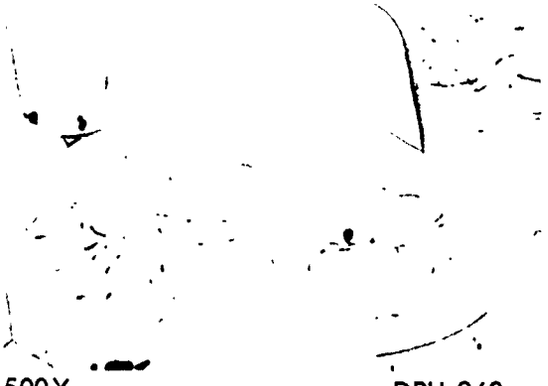
B. Swaged + 1 Hr/2552°F

500X DPH 354

C. Swaged + 1 Hr/2732°F

500X DPH 361

D. Swaged + 1 Hr/3000°F



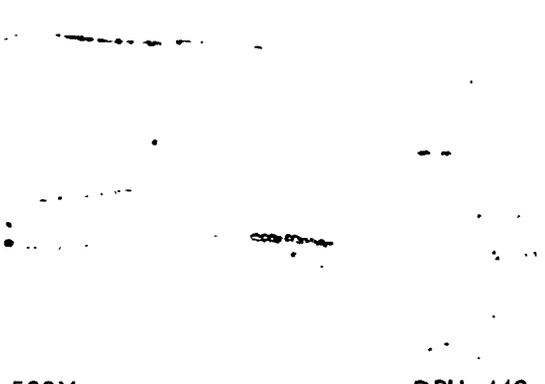
500X DPH 362

E. Swaged + 1 Hr/3272°F

500X DPH 370

F. Swaged + 1 Hr/3632°F

Figure 4. Microstructures of NASVF-200 (Ta-16W-1Re-0.7Hf-0.025C) Swaged and Heat Treated at Various Temperatures



500X DPH 442

A. As Swaged

500X DPH 374

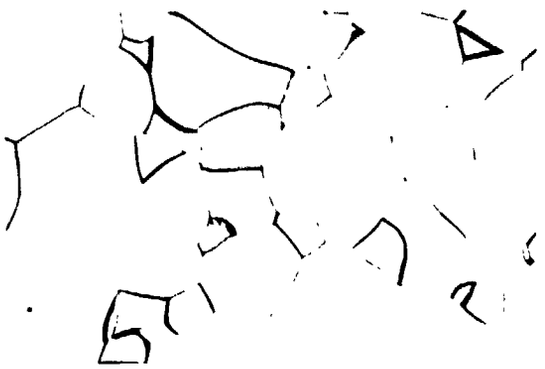
B. Swaged + 1 Hr/2552°F

500X DPH 382

C. Swaged + 1 Hr/2732°F

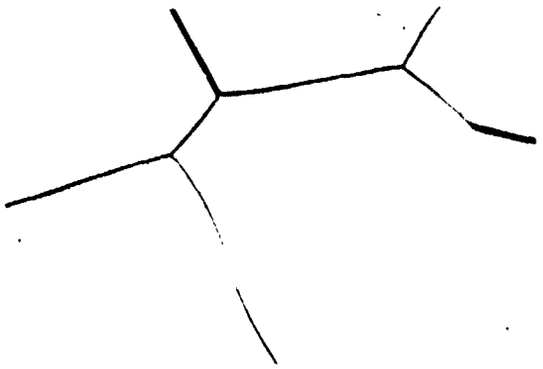
500X DPH 397

D. Swaged + 1 Hr/3000°F



500X DPH 395

E. Swaged + 1 Hr/3272°F



500X DPH 285

F. Swaged + 1 Hr/3632°F

Figure 5. Microstructures of NASVF-300 (Ta-14W-1Re-0.7Hf-0.015N-0.015C) Swaged and Heat Treated at Various Temperatures

the figures, complete recrystallization occurred in the temperature range 2700 to 3000°F. The 2550°F swaging temperature promoted recovery during reheat between swaging passes. As a result the temperature required for complete recrystallization is higher due to the lower strain energy in the warm worked material.

4.5 Mechanical Properties

4.5.1 Tensile Properties

Both notched and smooth bar tensile tests were conducted on the scale-up alloys and the test results are in Tables 6 and 7. Because of severe losses during swaging and machining of NASVF-200 and NASVF-300, a limited number of test specimens for these compositions were obtained. As a result, NASVF-300 was not tensile tested and only limited tensile testing of NASVF-200 was accomplished.

The smooth bar tensile strength data obtained on NASVF-100 and NASVF-200 was similar to that obtained on the corresponding screening compositions studied in Phase I. The strength properties represent a significant improvement over ASTAR-811C, the reference material. The room temperature tensile strength and ductility and 2400°F tensile strength of NASVF-100 and NASVF-200 are compared with ASTAR-811C in Figure 6. As can be seen in this figure, increasing the tungsten content from 8% in ASTAR-811C to the 14% level for NASVF-100 resulted in a significant increase in both room temperature and 2400°F tensile strength with negligible effect on room temperature ductility. However, increasing the tungsten to 16% (2% additional over NASVF-100) resulted in a modest increase in room temperature strength but the 2400°F tensile properties remained unchanged. Examination of the fractures of tested tensile specimens revealed that at 2200°F and below, failure occurred by transgranular shear while above this temperature fracture was almost entirely by intergranular separation. Examples of tensile fractures at 1800°F, 2200°F, and 2600°F as shown in Figure 7. Thus it would appear that the grain boundary strength is limiting elevated temperature tensile strength. The tensile ductility of NASVF-200 was significantly less than NASVF-100

Table 6. Tensile Data for Advanced Tantalum Base Alloys

Identification and Nominal Composition	Test Condition	Test Temperature °F	Yield Strength ksi	Ultimate Strength ksi	Elongation Uniform %	Elongation Total %	Reduction in Area %
NASVF-100 Ta-14W-1Re-0.7Hf-0.025C	As Swaged	RT	166.9	170.1	0.68	1.0	1.9
	Swaged plus 1 hr/2552°F	RT	135.8/131.8	141.7	14.9	30.0	65.3
	Swaged plus 1 hr/2912°F	RT	147.0/138.4	149.0	14.8	26.4	60.1
	Swaged plus 1 hr/3272°F	RT	.33.6/130.1	139.0	14.7	28.0	42.8
	Swaged plus 1 hr/3632°F	RT	137.4	137.4	6.3	6.4	8.3
	Swaged plus 1 hr/3272°F	1800	52.6	103.2	16.1	20.0	53.5
	Swaged plus 1 hr/3272°F	2200	50.5	89.0	16.2	21.0	60.3
	Swaged plus 1 hr/3272°F	2600	39.9	49.0	6.4	27.0	55.8
NASVF-200 Ta-16W-1Re-0.7Hf-0.025C	Swaged plus 1 hr/3272°F	RT	159.5/152.9	159.5	13.2	18.4	27.3
	Swaged plus 1 hr/3272°F	2400	45.9	65.4	7.1	10.0	41.2

Table 7. Notched-Strength Data for Advanced Tantalum Base Alloys

Identification and Nominal Composition	Test Condition	Test Temperature °F	Ultimate Strength Notched ksi	Ultimate Strength Unnotched ksi	Notched Strength Ratio
NASVF-100 Ta-14W-1Re-0.7Hf-0.025C	Swaged 1 hr/3272°F Swaged 1 hr/3272°F	RT -50	192.7 188.1	139.0	1.38
NASVF-200 Ta-16W-1Re-0.7Hf-0.025C	Swaged 1 hr/3272°F	RT	159.5	233.4	1.46

* $K_t = 2.9$

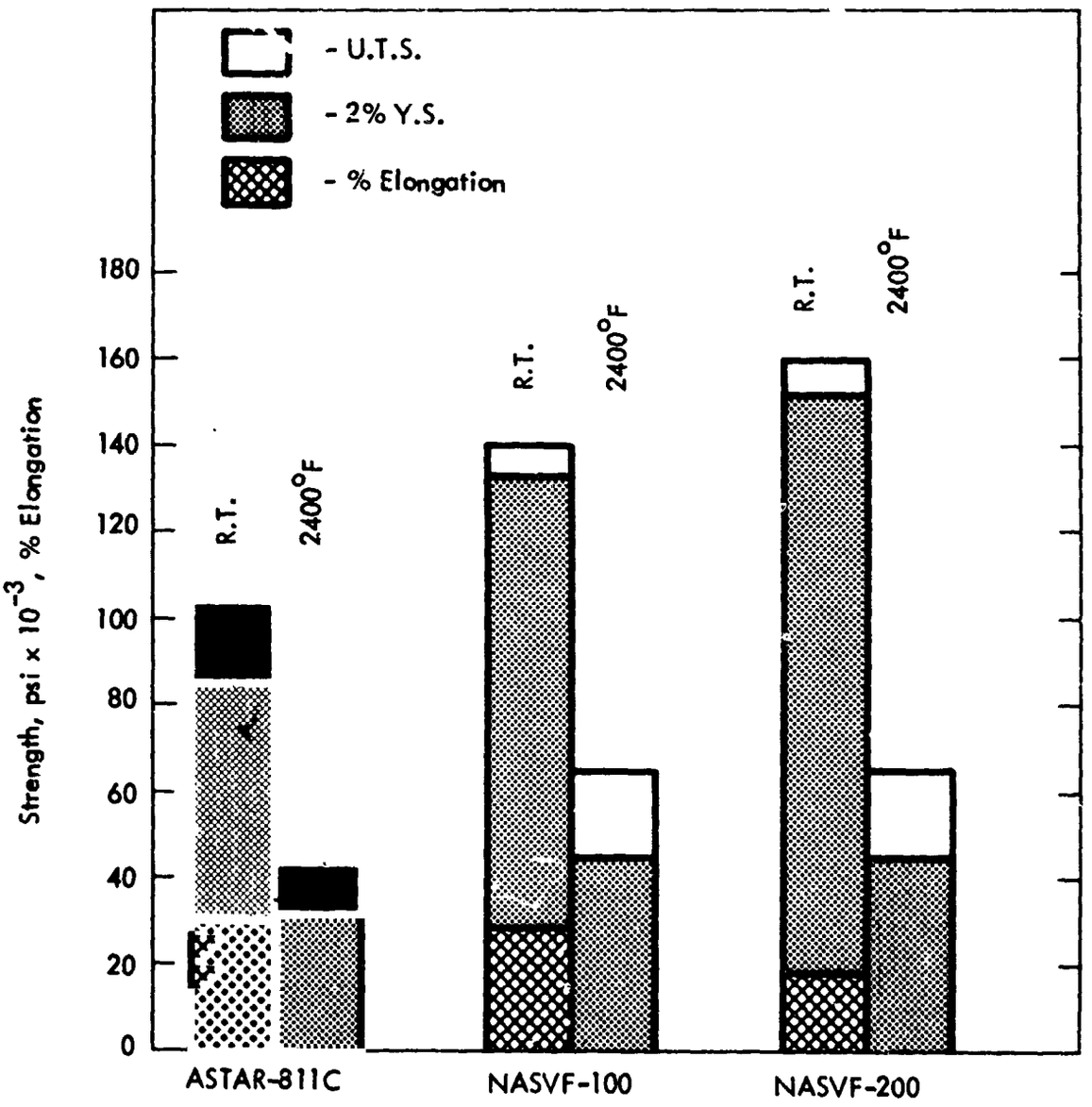


FIGURE 6 - Comparison of High Strength Tantalum Alloy Compositions with ASTAR-811C

Figure 7. Elevated Temperature Fracture Appearance of NASVF-100

(60% R.A. vs 27% R.A.). Thus the additional 2% tungsten in NASVF-200 resulted in degradation of low temperature ductility without providing any elevated temperature tensile strength advantage.

As is evident from the data in Table 6, both NASVF-100 and NASVF-200 were both notch insensitive on the basis of the ratio of the notched to unnotched tensile strength.

Room temperature tensile strength and ductility was definitely influenced by the final annealing temperature. The data in Table 6 are represented graphically in Figure 8. Here it can be seen that annealing for an hour at 2550°F (1400°C) resulted in a significant improvement in room temperature ductility accompanied by a moderate decrease in tensile strength. Very little change in R.T. tensile strength was exhibited after annealing at temperatures up to 3630°F (2000°C). However, room temperature ductility was severely reduced after annealing at 3630°F (2000°C).

4.5.2 Creep Properties

Temperature change and constant load creep tests were performed on all three scale-up alloy compositions. The test data are in Table 8 and presented graphically in Figure 9. Specimens were annealed for one hour at 3300°Fprior to testing since preliminary experimentation on NASVF-100 indicated that a one hour anneal at 3600°F did not provide a significant improvement in creep properties over a 3300°F recrystallization treatment (See Figure 9).

Both 3300°F and 3600°F annealed material had better creep properties than did the material annealed at 3000°F (See Figure 9). This behavior is different than exhibited by ASTAR-811C[1] where is was demonstrated that annealing at 3600°F resulted in clearly superior creep properties over material recrystallized at 3300°F and below. Since the 3600°F anneal did not result in significantly better creep properties and did cause a severe loss in room temperature ductility (See Figure 8), a final annealing treatment of one hour at 3300°F was selected.

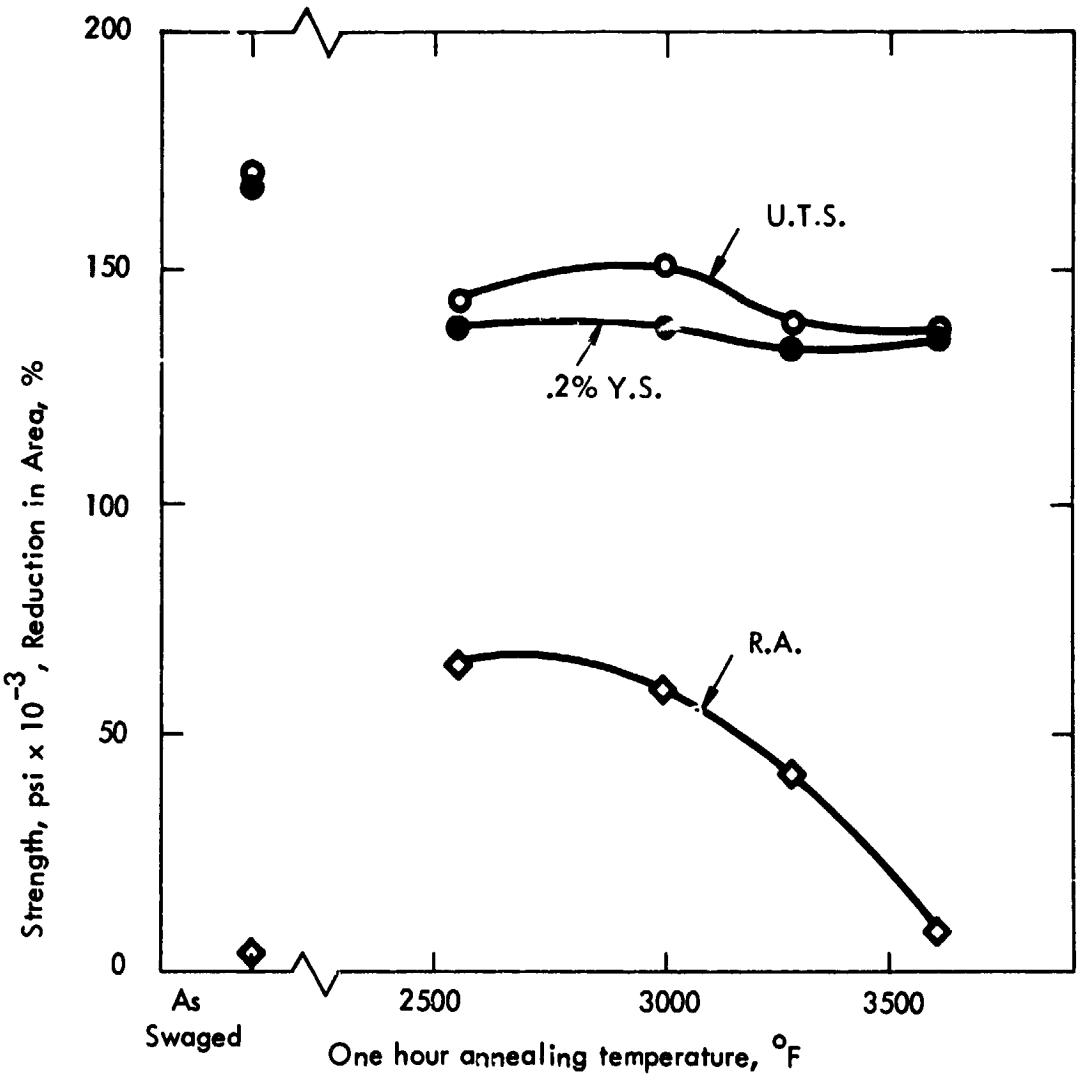


FIGURE 8 - Effect of Annealing Temperature on Room Temperature Strength and Ductility of Ta-14W-1Re-0.7Hf-0.025C

Table 8. Creep Properties of Advanced Tantalum Base Alloys (Swaged Rod)

Specimen Identification and Condition	Stress (ksi)	Test Temperature (°F)	Strain on Loading (%)	Primary Creep Strain (%)	Primary Creep Time (hrs)	Secondary Creep Strain (%)	Secondary Creep Time (hrs)	Secondary Creep Rate (%/hr)	Time to 1% Strain (hrs) (2)	L-M Parameter(1) $T_{O_R}(15+\log t) \times 10^{-3}$
NASVF-100 Final Anneal 1 hr/3000°F	50	1850	0.5	1.2	70	0.88	167	0.00525	190	39.9
	50	1800	(4)	--	--	0.23	500	0.00046	2174	41.4
	30	2250	0.15	0.15	2	2.65	89	0.0298	34	44.8
	30	2200	(4)	--	--	2.68	180	0.0148	68	44.8
NASVF-100 Final Anneal 1 hr/3600°F	50	1850	0.71	1.64	220	0.13	110	0.0012	833	41.4
	50	1800	(4)	--	--	0.29	600	0.0008	2083	41.4
	30	2300	0.08	0.11	2	3.05	112	0.0272	37	45.7
	30	2250	(4)	--	--	2.52	223	0.0113	88	45.9
	30	2200	(4)	--	--	1.28	190	0.0067	147	45.7
NASVF-100 Final Anneal 1 hr/3300°F	50	1850	0.41	0.74	27	0.88	193	0.0045	222	40.1
	50	1800	(4)	--	--	0.28	616	0.00045	2222	41.5
	40	2000	0.13	0.87	100	1.45	755	0.00192	520	43.5
	40(3)	2000	0.14	0.00	00	2.00	1432	0.00140	714	43.9
	20	2300	0.07	0.00	00	0.61	1634	0.00037	2703	50.9
	20	2400	0.16	0.00	00	2.17	847	0.00256	390	50.3
NASVF-200 Final Anneal 1 hr/3300°F	40	2000	0.30	0.35	73	0.13	1102	0.00012	8333	46.5
	40	2100	(4)	--	--	1.23	864	0.0014	714	45.7
	20	2400	0.10	0.00	00	1.20	1247	0.00096	1042	51.5
	20	2300	0.10	0.00	00	0.12	791	0.00015	6667	51.9
	20	2350	(4)	--	--	0.39	1016	0.00038	2605	51.8
	15	2500	0.04	0.00	00	0.97	1061	0.00091	1100	53.4
NASVF-300 Final Anneal 1 hr/3300°F	27.5	2300	0.14	0.00	00	0.00	353	--	--	--
	27.5	2350	(4)	0.00	00	0.00	241	--	--	--
	27.5	2400	(4)	0.00	00	1.89	561	0.0034	296	50.0
	27.5	2350	(4)	0.00	00	0.87	291	0.0030	334	49.2
	27.5	2300	(4)	0.00	00	0.45	318	0.00142	704	49.3
	40	2000	.24	0.00	00	0.17	690	0.00025	4000	45.8
	40	2100	(4)	0.00	00	.78	943	0.00083	1205	46.3

(1) Larson Miller Parameter = T_{O_R} (15 + log t) where t is time in hours to creep 1%

(2) Calculated from secondary creep rate

(3) Specimen st ·ed 10% at 2000°F in tension test prior to creep testing

(4) $\Delta\sigma - \Delta T$ creep test

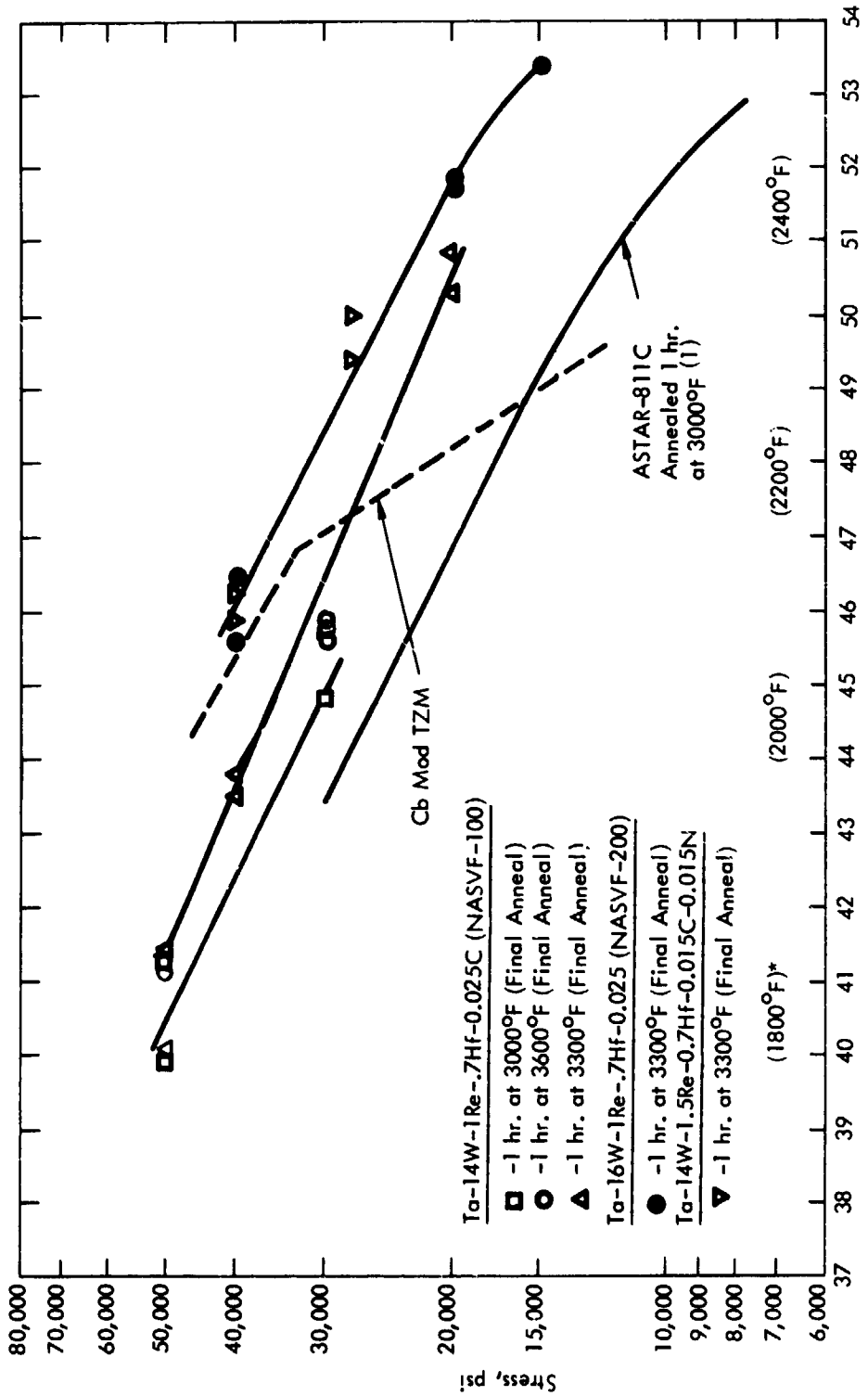


FIGURE 9 - Creep Properties of Experimental Tantalum Base Alloys

* Temperature for 1% creep in 1000 hours

The creep properties of NASVF-200 were clearly better than NASVF-100 (See Figure 9). Thus, the higher tungsten content does provide enhancement of time dependent properties, although no benefit was shown for the time dependent elevated temperature tensile strength. The carbonitride strengthened NASVF-300 exhibited creep properties equivalent to a slightly superior to NASVF-200. NASVF-300 has the same tungsten content as NASVF-100 but has approximately half of the carbon substituted with nitrogen. The effectiveness of the carbonitride precipitate has been demonstrated previously[1]. However, probable liquid alkali metal compatibility of the nitride may preclude its usage in liquid alkali metal systems. However, the carbonitride phase is definitely superior to the carbide at temperatures below 2400°F. From Figure 9 it is also evident that the advanced high strength tantalum alloys are superior to the high strength molybdenum alloy Cb-TZM for long time usage above about 2100°F.

Various shaped creep curves were observed during the course of testing the advanced high strength tantalum alloys and typical examples for both carbide strengthened compositions are shown in Figures 10 and 11. At 2300°F and above, the curves are characterized by a loading strain, no transient creep and either fairly linear or a perceptible constantly increasing rate which results in a concave upward curve. (See Figure 10b and d and Figure 11a, b and d). At 2000°F, the curves typically consists of a loading strain, a rather large transient strain followed by a definite secondary stage (See Figure 11c) and if the test was continued for a sufficient period, entry into third stage (Figure 10a). The large transient strain observed for NASVF-100 (See Figure 10a) was eliminated by straining the specimen 10% by a uniaxial tensile test at 2000°F prior to creep testing. The creep curve of the pre-strained specimen exhibited a similar loading strain to the unstrained specimen but primary creep was eliminated. The secondary creep rate was about the same in both cases (See Table 7 and Figure 10a and c).

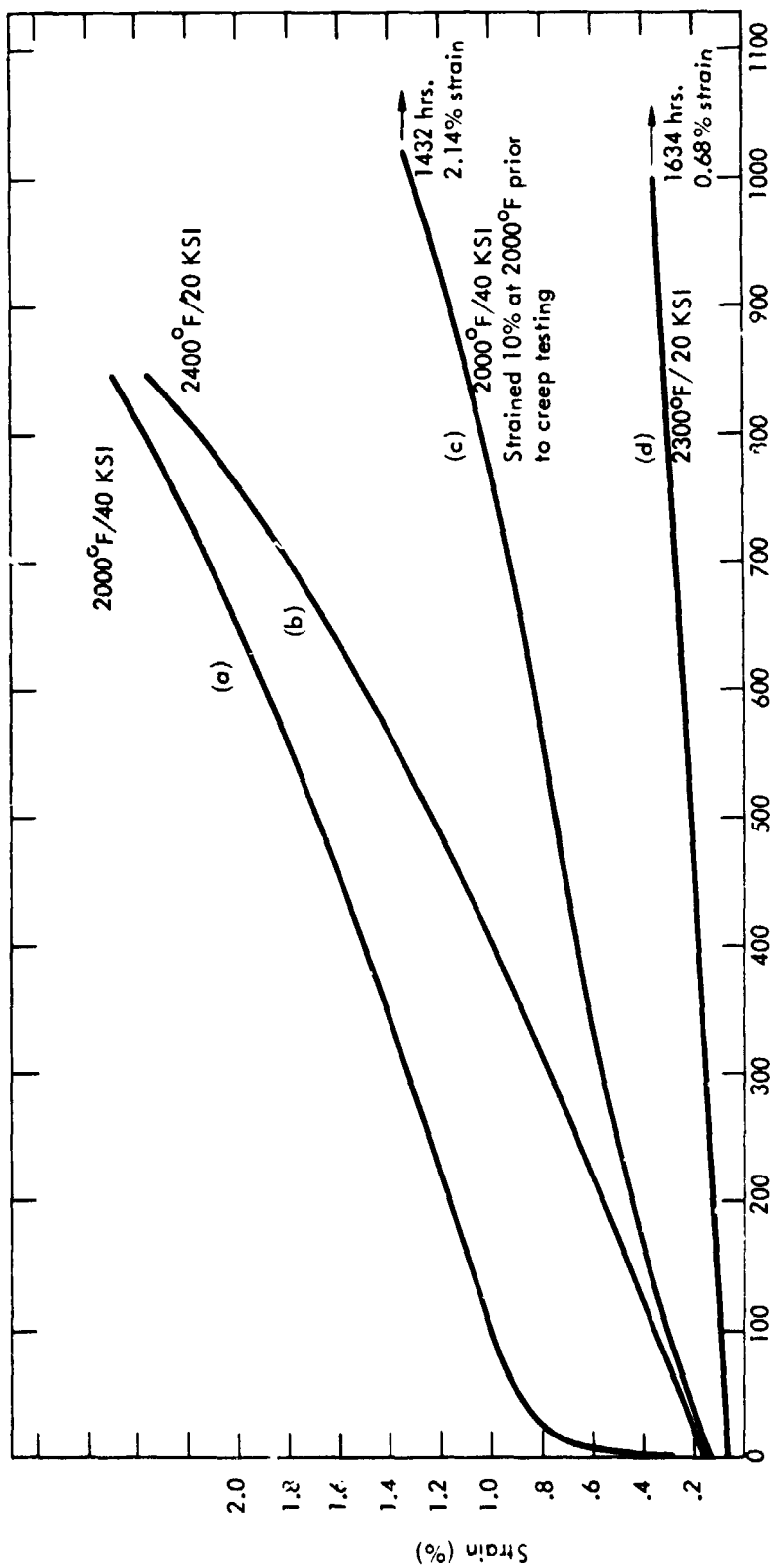


FIGURE 10 - Creep Curves for Tantalum Alloy NASVF 100

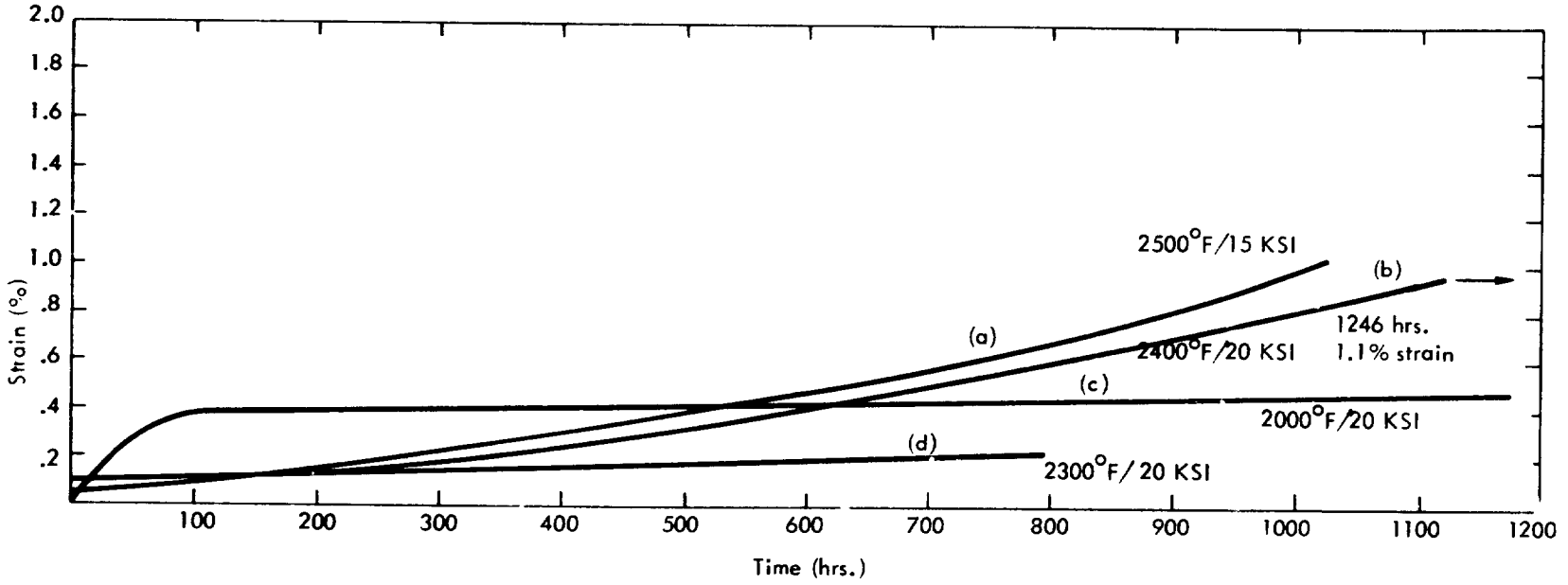
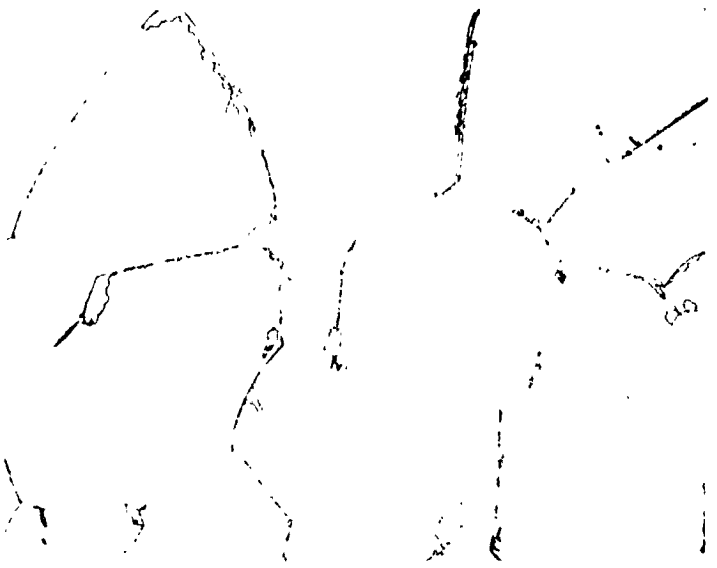


FIGURE 11 - Creep Curves for Tantalum Alloy NASVF-200

The post-test microstructure of both specimens are shown in Figure 12. Large blocky grain boundary carbide phase is present in both specimens. However, the pre-strained specimen contains a precipitate distributed throughout the matrix where the matrix of the unstrained matrix appears to be relatively free of precipitates. It is hypothesized that the dislocations generated during the prestraining acted as nucleating sides for subsequent carbide precipitation. Although there is a difference in microstructure of the two specimens, the identical secondary creep rates observed would tend to indicate that observable carbide precipitate distribution had little effect on the creep behavior. Thus elimination of the primary creep occurred as a result of the strain energy introduced during the pre-straining.

Generally, the microstructures of the carbide strengthened alloys consist of a Ta_2C precipitate dispersed in the single phase matrix as illustrated by the photomicrographs shown in Figure 13. Microstructures similar to this have been observed in ASTAR-811C as well as during the Screening Phase Investigation[4]. Also during the screening phase investigation, the thermal instability of the carbide precipitate was observed and prompts the question as to the role of the carbide in improving creep strength of tantalum base alloys. It is well established that the Ta_2C carbides that precipitate are non-coherent, and that the precipitation kinetics are rapid[1,4]. This is also illustrated by the upper curve plotted in Figure 14 which is a plot of the room temperature hardness as a function of aging time for a Ta-16W-2Re-0.7Hf-0.025C (NASVF-2). The lower curve is the creep curve observed for NASVF-200, Ta-16W-1Re-0.7Hf-0.025C, tested at 2000°F and 40,000 psi. Interestingly, the end of primary creep coincides with minima in the room temperature hardness plot. Thus the carbide strengthened tantalum alloys are undergoing precipitation softening, at least as measured by room temperature hardness. The exact mechanism whereby the carbide or carbon enhances elevated temperature creep strength is unclear, but the effect of carbon on the creep strength has been well demonstrated[1]. Creep curves of ASTAR-811C and a carbon free modification (ASTAR-811) shown in Figure 15 show that carbon is indeed an effective strengthener.

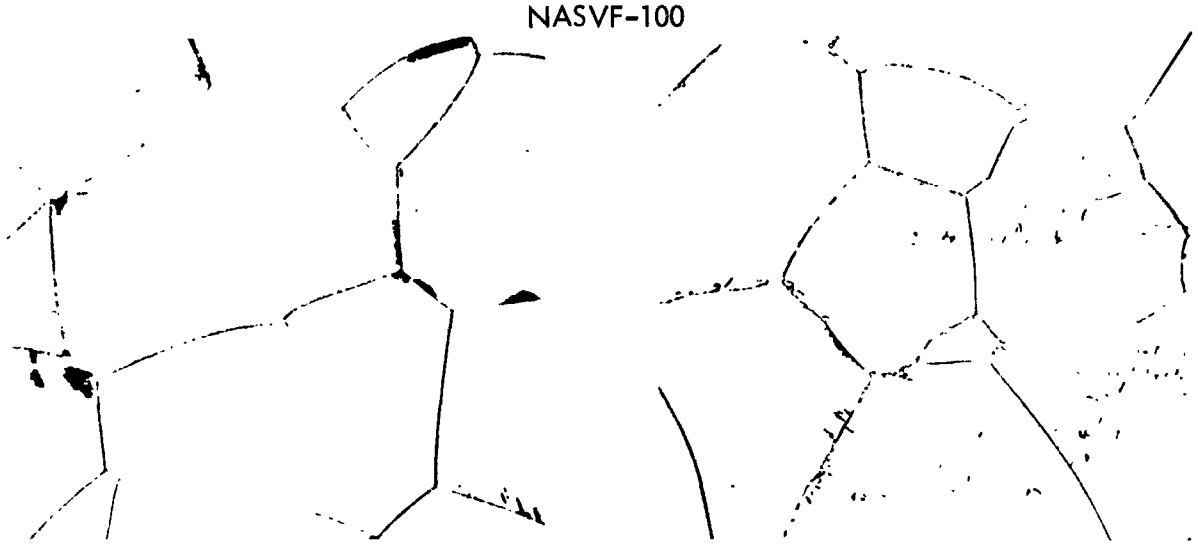


(a) Annealed One Hour at 3300°F, then Creep Tested at 2000°F and 40,000 psi
500X

(b) Same as (a) Except Strained 10% at 2000°F Prior to Creep Testing
500X

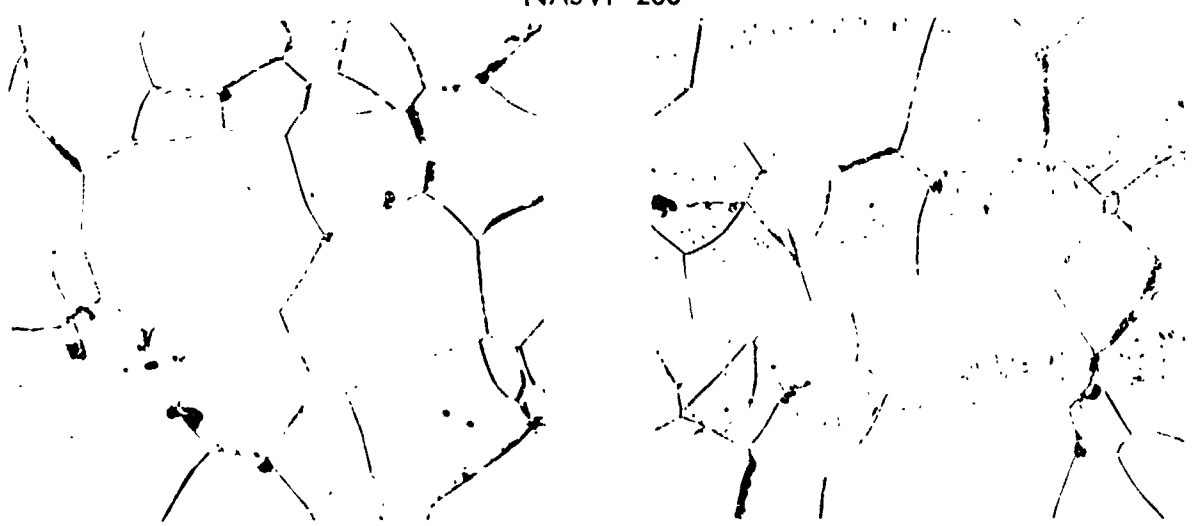
Figure 12. Influence of Pre-Strain on the Microstructure of NASVF-100 Creep Tested at 2000°F and 40,000 psi

NASVF-100



a) tested 2300°F and 20,000 psi for 1,634 hrs b) 2400°F and 20,000 psi for 847 hours

NASVF-200



c) tested 2400°F and 20,000 psi for 1,247 hrs d) tested 2500°F and 15,000 psi for 1,061 hrs

Figure 13. Microstructures of NASVF-100 and NASVF-200 After Various Creep Test Exposures. All Specimens Annealed One Hour at 3300°F Prior to Test 500X

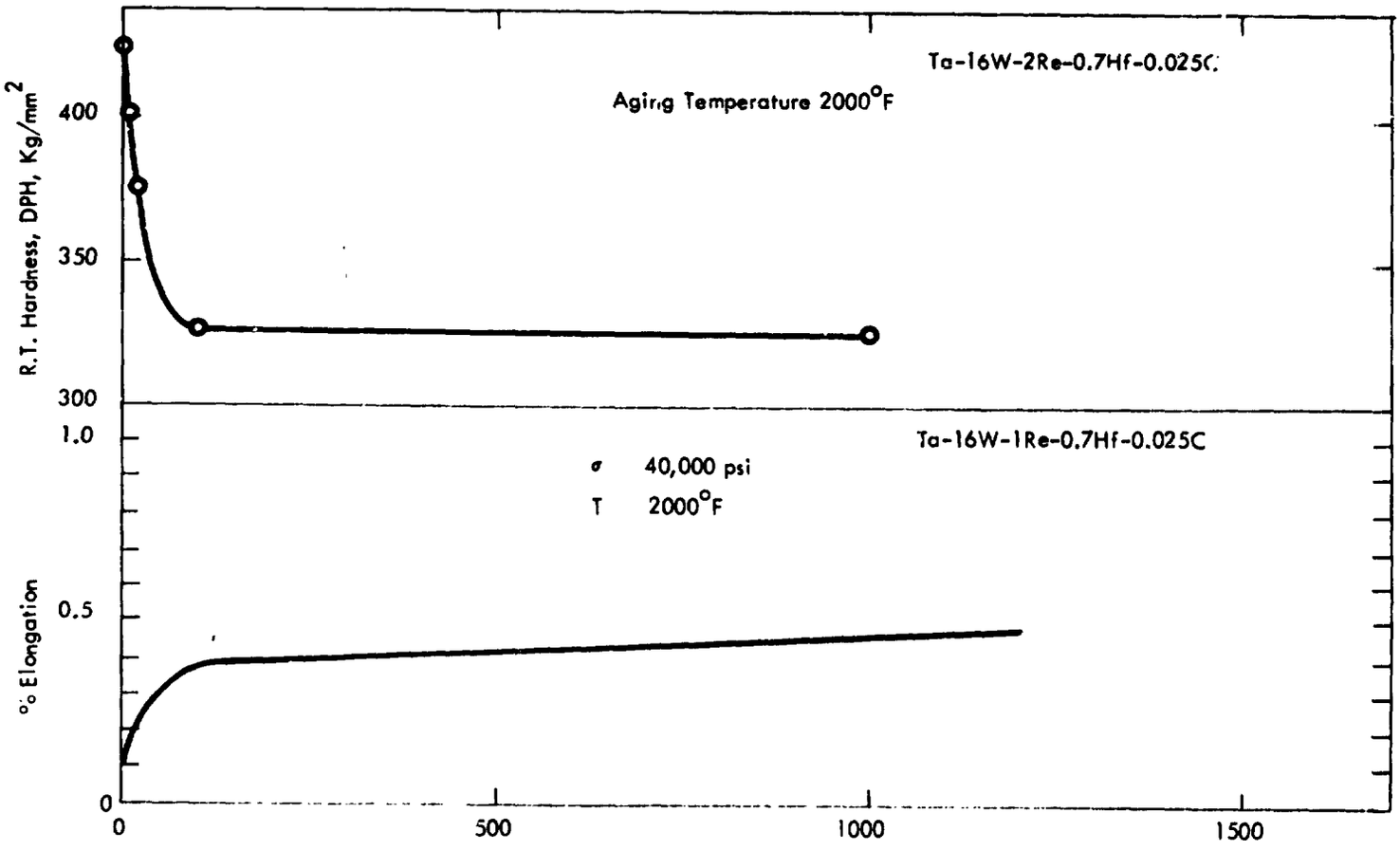


FIGURE 14. Aging Response and Creep Curve for Carbide Strengthened Tantalum Alloy

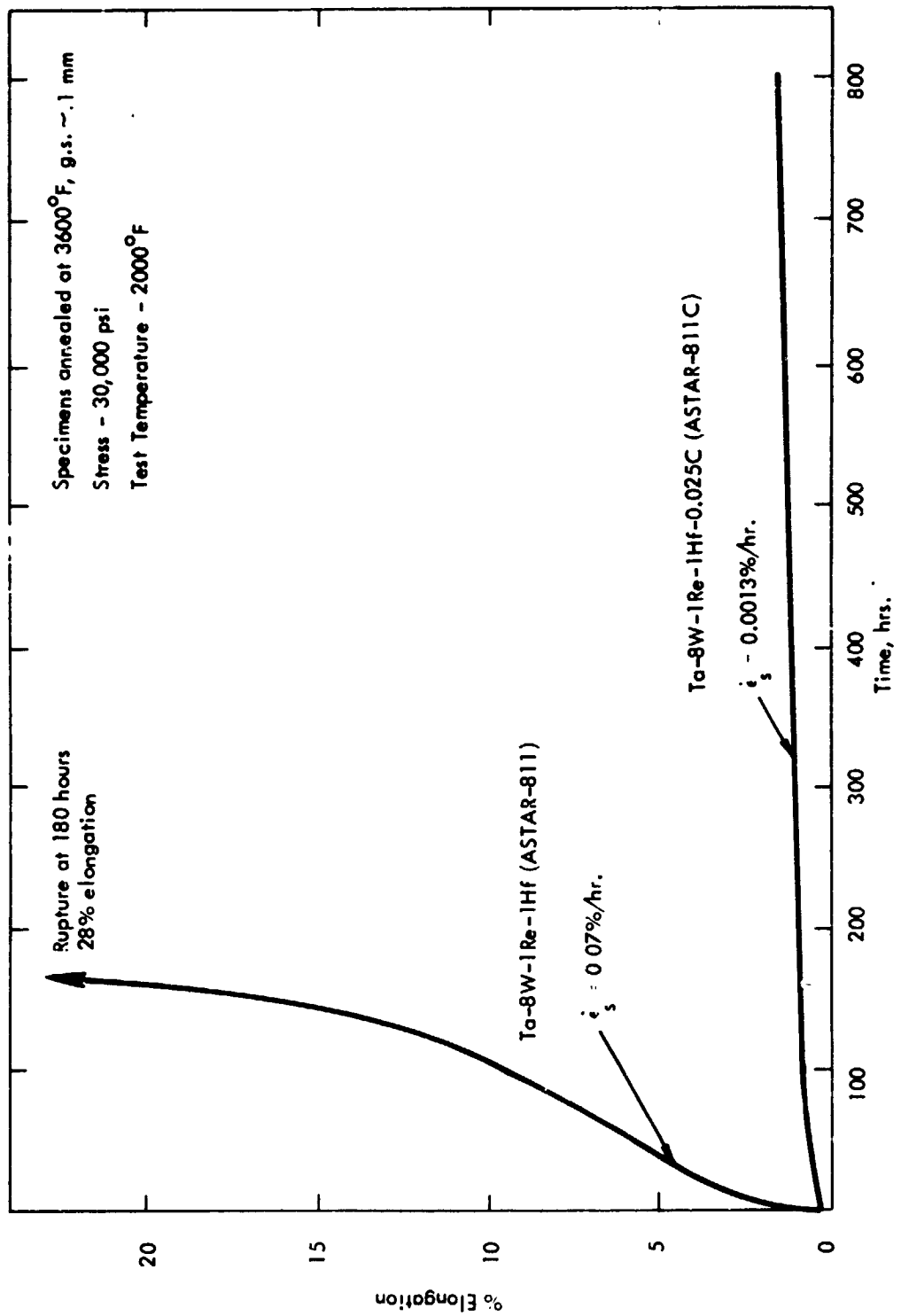


Figure 15. Effect of Carbon on Creep Behavior of ASTAR-811C at 2000°F and 30,000 psi

4.6 Forging Evaluation

The forgeability of the three alloys was evaluated at three elevated temperatures and at two reduction ratios at each temperature. Forging was done on a Dynapak - high energy rate machine. Forging billets were cut from the extruded bar stock, annealed one hour at 3000°F, plasma coated with molybdenum, heated to the forging temperature in an inert environment induction heater, and then forged in one stroke in the Dynapak. Forging data are given in Table 9. To achieve the desired reduction ratios, a spacer plate of the appropriate thickness was used to stop the moving platen of the Dynapak. Target reduction ratios of 2:1 and 3:1 at forging temperatures of 2200, 2500 and 2800°F were investigated to assess the forging characteristics of the three advanced tantalum alloys. The "as forged" discs are shown in Figures 16, 17, and 18. All three alloy billets forged without catastrophic failure. Except for some minor edge cracking, the alloys appear to be quite amenable to forging. The forgeability also appears to be independent of forging temperature over the range investigated. In each case the forgings, which experienced the greatest reduction, exhibited the greatest degree of edge cracking.

In Figure 19, the cross sections of two forged discs of Ta-14W-1Re-0.7Hf-0.025C (NASVF-100) are shown. The sample forged at 2800°F at a 2:1 reduction exhibits a typical wrought structure of an up-set forged material. The sides are essentially crack free and the overall appearance attests to the forgeability of the material under the given conditions. The sample forged at 2500°F at a 3:1 reduction also exhibits a typical macrostructure of an up-set forged material. The cracks which are evident at the side of the forging are shown to be superficial. All three alloy compositions responded to the forging operation in a similar manner.

4.7 Welding Evaluation

A cursory welding evaluation was conducted to determine the adaptability of the three alloy compositions to a specialized EB welding application. It was recognized that the particular alloys under investigation in this program are being developed primarily as forging alloys where a high degree of weldability, although desirable, is not necessary. It has been

Table 9. Forging Data for Advanced Tantalum Alloys

Billet Identification and Size	Forging Temperature (°F)	Forged Dimensions Height (in)	Dia. (in)	Reduction Ratio	
NASVF-100	2200	0.403	1-1/2	2.4:1	a
Ta-14W-1Re-0.7Hf-	2200	0.338	1-1/2	2.8:1	b
0.025C	2500	0.362	1-1/2	2.6:1	a
D_o = 0.95"	2500	0.320	1-5/8	3.0:1	b
H_o = 0.95"	2800	0.477	1-1/4	2.0:1	a
	2800	0.291	1-5/8	3.3:1	b
NASVF-200	2200	0.440	1	1.8:1	c
Ta-16W-1Re-0.7Hf-	2200	0.370	1	2.2:1	d
0.025C	2500	0.512	1	1.6:1	c
D_o = 0.80"	2500	0.370	1	2.2:1	d
H_o = 0.85"	2800	0.492	1	1.6:1	c
	2800	0.370	1	2.2:1	d
NASVF-300	2200	0.530	1-1/4	1.5:1	c
Ta-14W-1.5Re-0.7Hf-	2200	0.363	1-1/4	2.2:1	d
0.025C	2500	0.517	1-1/4	1.5:1	c
D_o = 0.80"	2500	0.358	1-1/4	2.2:1	d
H_o = 0.85"	2800	0.509	1-1/4	1.6:1	c
	2800	0.362	1-1/4	2.2:1	d

D_o Starting Billet Diameter

H_o Starting Billet Height

Spacer Size

a - 0.470
b - 0.310
c - 0.500
d - 0.330

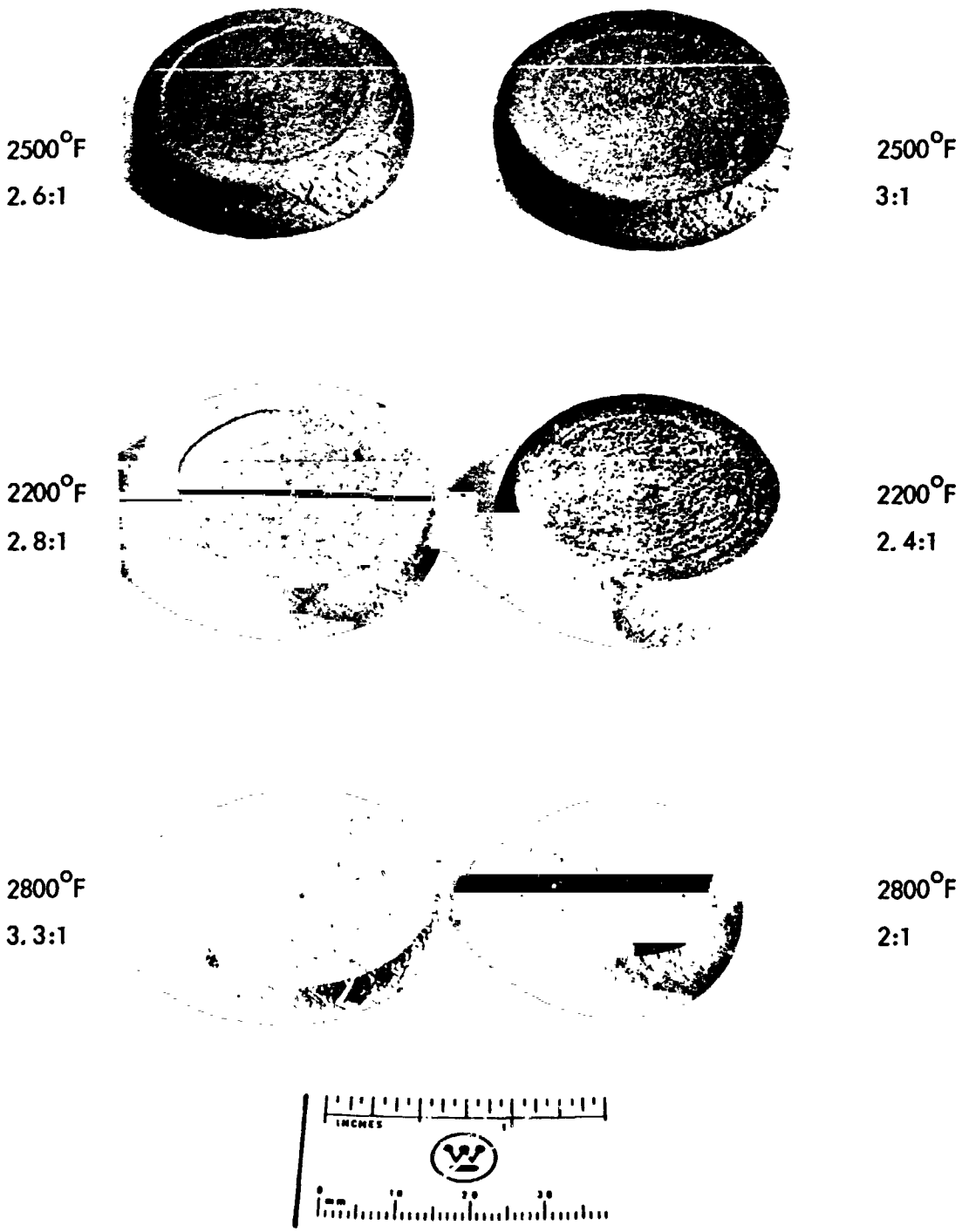


Figure 16. Upset Forged Specimens of Ta-14W-1Re-0.7Hf-0.025C (NASVF-100A) Forging Temperature °F/Reduction Ratio

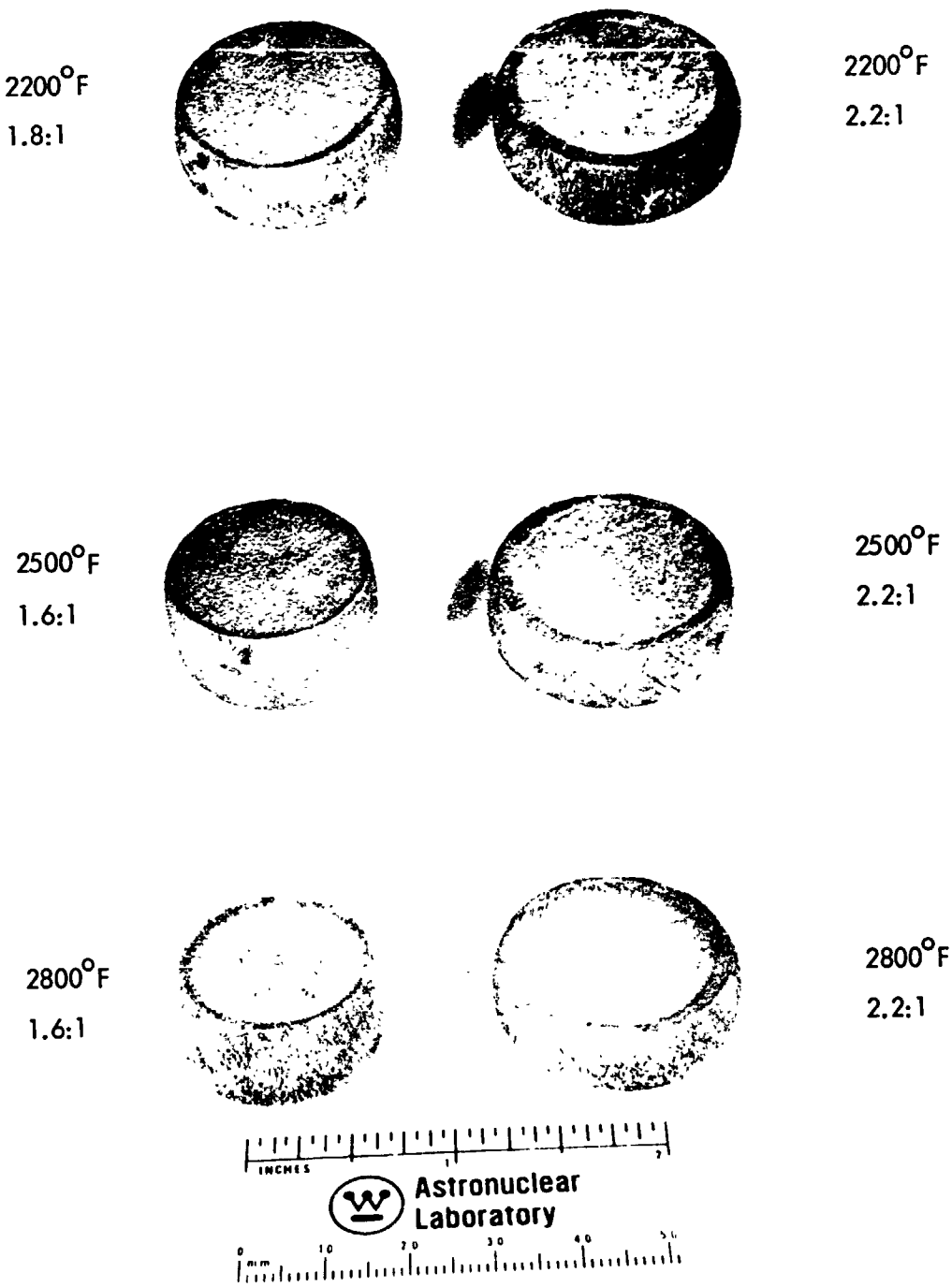


Figure 17. Upset Forged Specimens of Ta-16W-1Re-0.7Hf-0.025C (NASVF-200A) Forging Temperature °F/Reduction Ratio

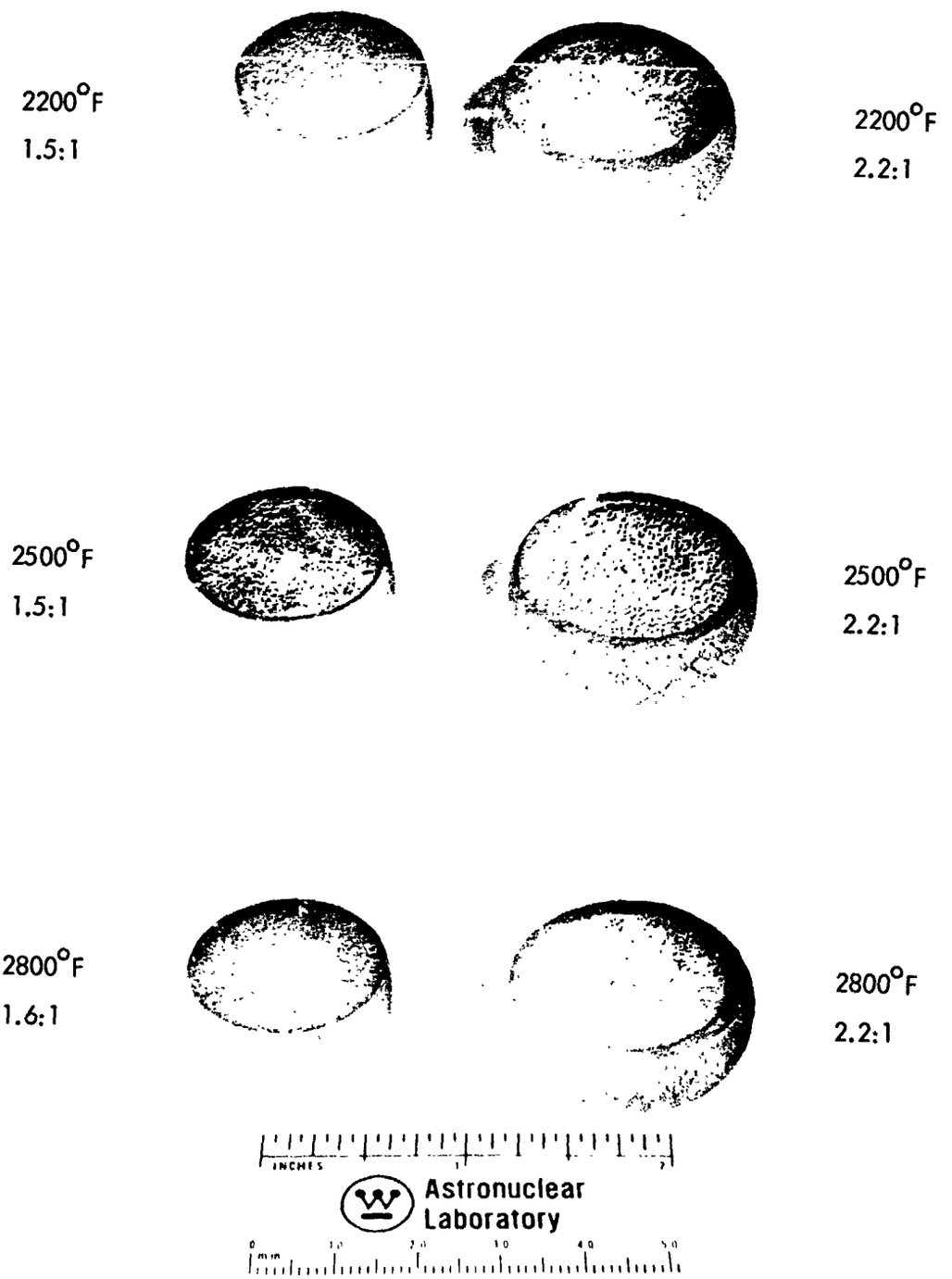


Figure 18. Upset Forged Specimens of Ta-14W-1.5Re-0.7Hf-0.015C-0.015N (NASVF-300A) Forging Temperature °F/Reduction Ratio

(a) Upset at 2800°F, Reduction Ratio 2:1

(b) Upset at 2500°F, Reduction Ratio 3:1

Figure 19. Photomacrograph of Upset Forging Section Ta-14W-1Re-0.7Hf-0.025C (NASVF-100) Mag. 2.5X

demonstrated in numerous studies that in general an increase in solute content of a tantalum base alloy tends to raise the DBTT. This effect on DBTT is most noticeable in welded material. Weld bend ductile-to-brittle transition-temperatures tend to be higher when compared to base metal.

In this study, a mock-up of a weld joint suitable for EB welding (See Figure 20 for schematic drawing) was constructed of each alloy composition. The ring was machined from one of the forged discs. The smaller diameter bar was machined from the original extrusion. Both pieces were vacuum annealed one hour at 3000°F prior to EB welding. To provide a basis for comparison, a joint mock-up of TZM was included in the evaluation. To set welding parameters a TZM and a NASVF-100 sample were welded and sectioned. The cross sections are shown in Figure 21. The original plan was to EB weld the end joint first, penetrating into the machined void in the center bar. Then weld the "Tee" joint on the other side of the forged ring, also penetrating into the machined void of the center bar. The first welds were made successfully, however, during the subsequent "Tee" weld, thermal stresses caused the end weld to crack as shown in Figure 22. Both alloys failed in a similar manner. The crack which propagated through the axis of the weld also extended around the circumference of the joint. It was concluded that the multiple pass weld imposed too severe a thermal-stress load on the initial weld and therefore could be omitted.

All three tantalum alloys and the TZM samples were successfully end welded only. The results are shown in Figures 23, 24, and 25. Each alloy in the "as-welded" condition is shown in Figure 23, the as-welded condition with dye penetrant developer in place in Figure 24. All four alloys have a fault free appearance. The samples were surface ground in preparation for further examination. The purpose of the grinding operation was to remove extraneous material from the weld region. The removal of the material from the TZM and NASVF-100 alloys had no ill effect. However, the NASVF-200 and NASVF-300 failed by cracking as a result of the removal of a thin surface layer. The cracks are evident in

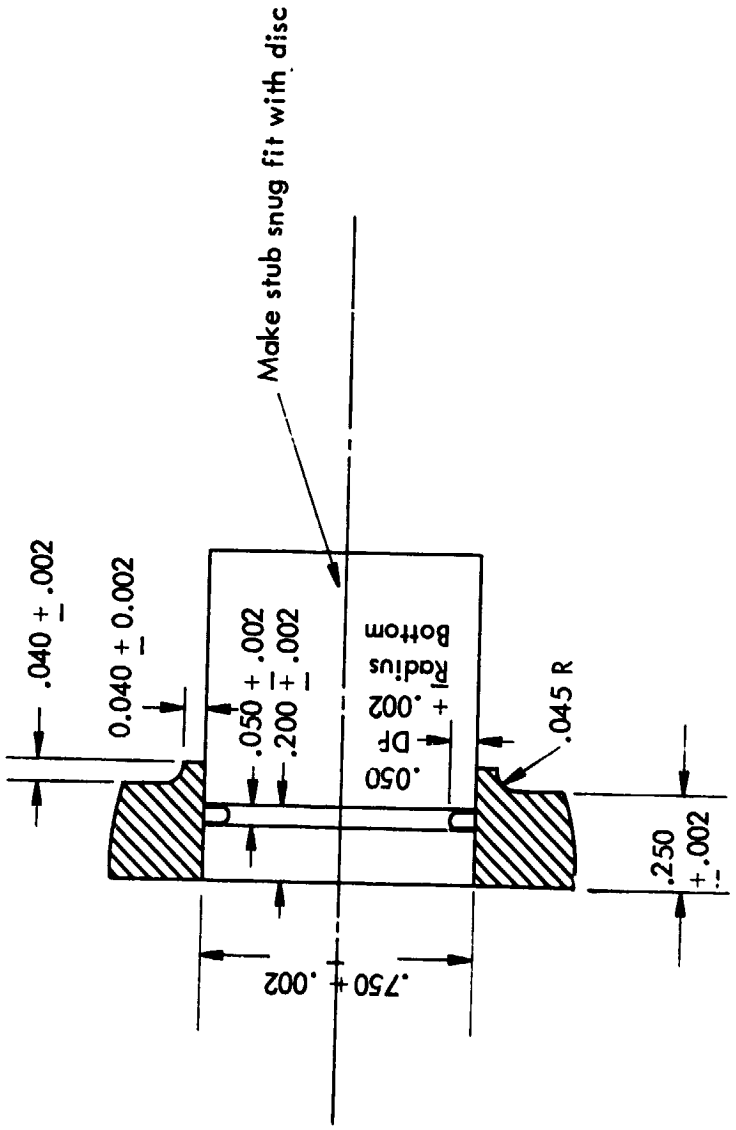


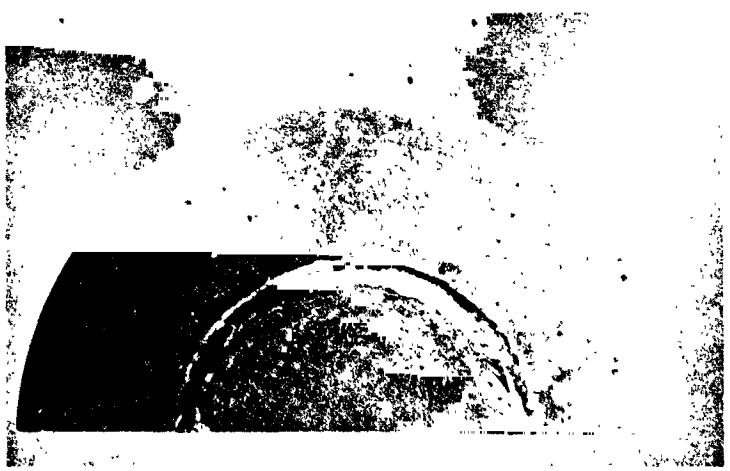
FIGURE 20 - EB Welding Sample

TZM

NASVF-100 4-1/2X

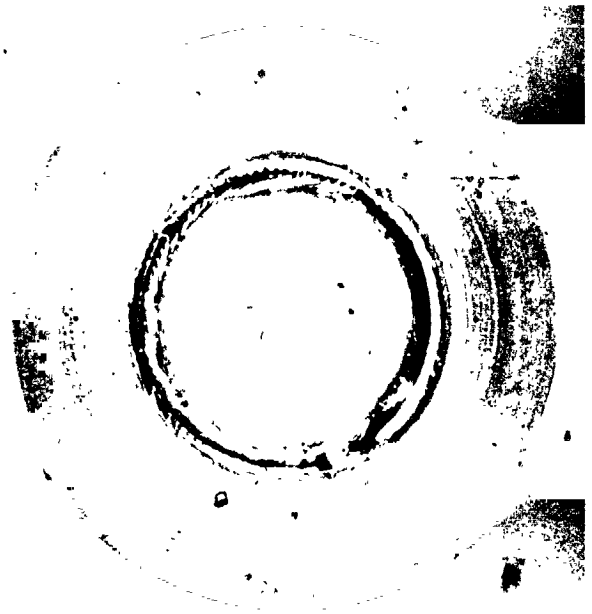
Figure 21. Cross-Section of TZM and NASVF-100 Weld Specimens

TZM



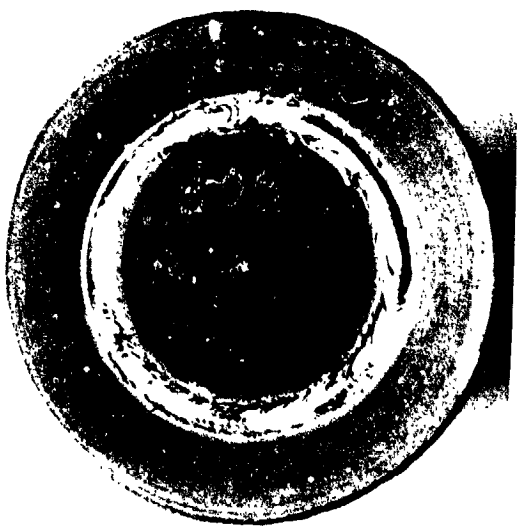
NASVF-100

Figure 22. Circumferential Cracks in NASVF-100 and TZM Caused by "Tee" Weld

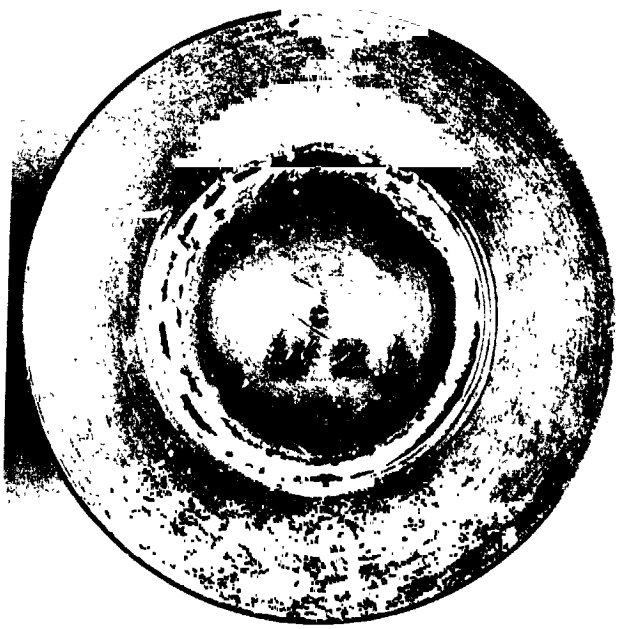


(a) NASVF-100

(b) NASVF-200

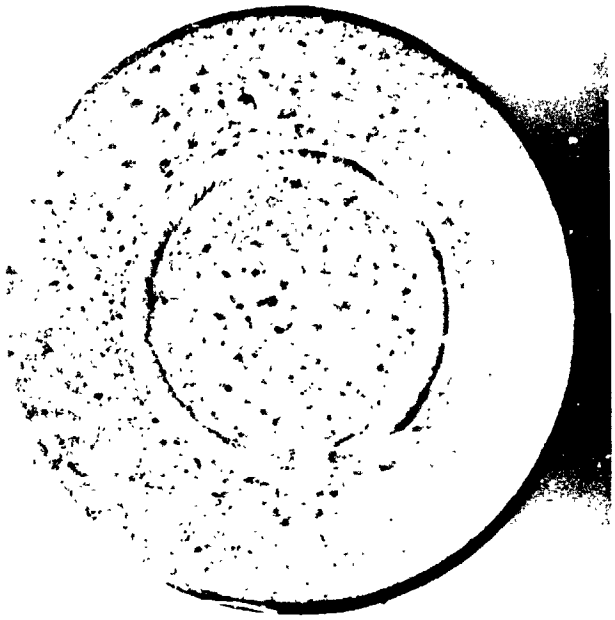


(c) NASVF-300

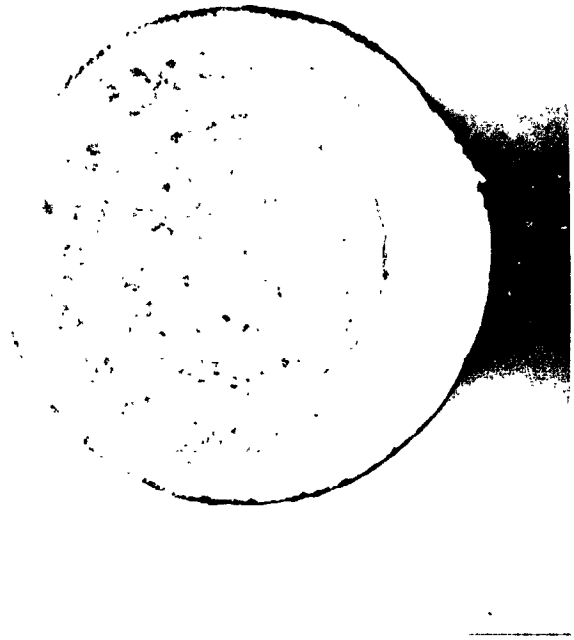


(d) Mo-TZM

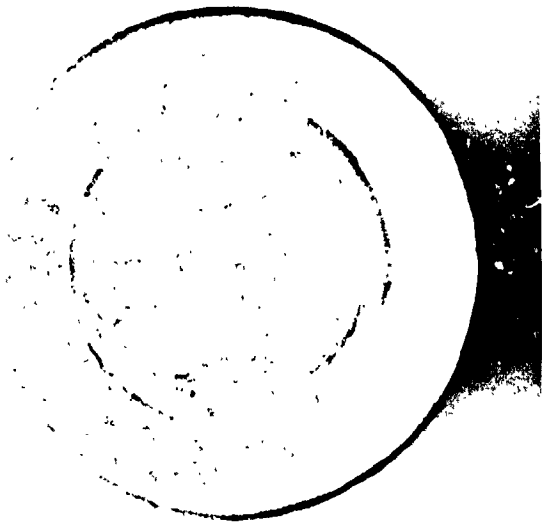
Figure 23. As-Welded Specimens of TZM and the Scale-Up Tantalum Base Alloys



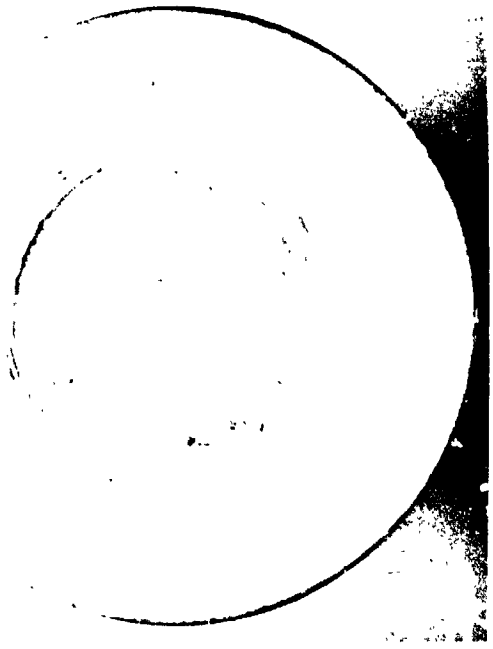
(a) NASVF-100



(b) NASVF-200

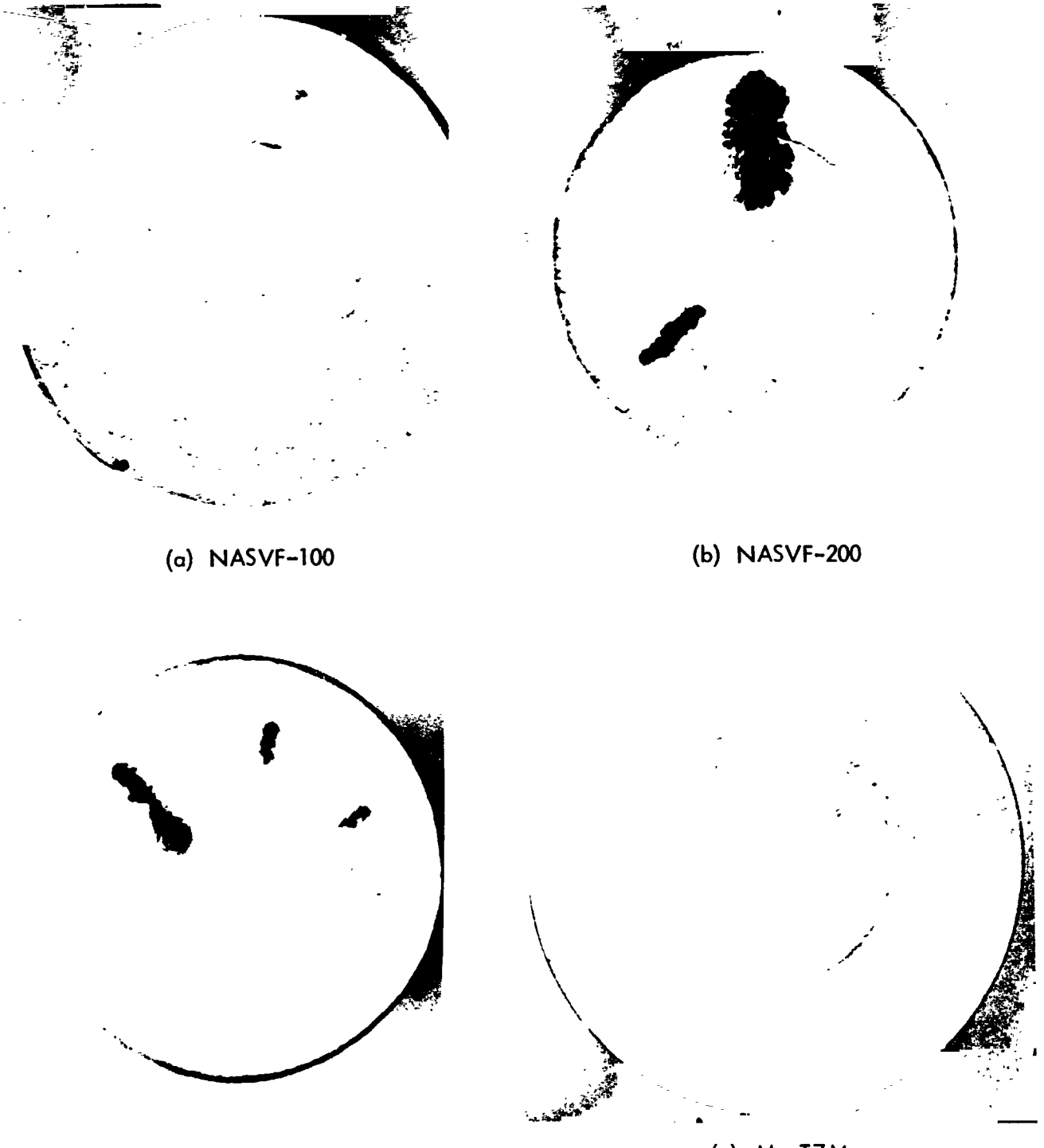


(c) NASVF-300



(d) Mo-TZM

Figure 24. As-Welded Specimens of TZM and Scale-up Tantalum Base Alloys with Dye Penetrant Developer in Place - Note absense of any indications



(a) NASVF-100

(b) NASVF-200

(c) NASVF-300

(d) Mo-TZM

Figure 25. Surface Ground End Welds Illustrating Defect Free NASVF-100 and Mo-TZM and Radial Cracks in NASVF-200 and NASVF-300

Figures 25b and c. The samples were not given a post weld anneal which undoubtedly would have prevented the failure. In both cases, the cracks are radial in nature and occurred in response to the high tensile hoop stress in the weldment. A stress relief anneal would have minimized the probability of this type of failure. The fact that the TZM and NASVF-100 alloys survived does indicate that a degree of weldability was achieved in those alloys.

5.0 CONCLUSIONS

Based on the results of this investigation, the scale-up compositions exhibited properties within the range predicted from the phase 1 screening investigation described in NASA CR-120818. Thus it was verified that high strength tantalum alloys still having good room temperature ductility could be prepared by increasing the tungsten content to the 14-16% level. The role of the carbide precipitate in enhancing high temperature creep strength however was not explainable.

Additional conclusions which were drawn from the scale-up investigation include the following:

(1) The carbonitride precipitate is a more effective creep strengthener than the straight Ta_2C precipitate.

(2) All three alloy compositions could be satisfactorily welded by the electron beam process.

6.0 REFERENCES

1) R. W. Buckman, Jr. and R. C. Goodspeed, "Development of Precipitation Strengthened Tantalum Base Alloys," WANL-PR-Q-017.

2) J. C. Sawyer and E. A. Steigerwald, "Generation of Long Time Creep Data on Refractory Alloys at Elevated Temperature," Final Report No. ER-7203, TRW Inc. Cleveland, Ohio, June 6, 1967.

3) R. W. Buckman, Jr. and R. C. Goodspeed, "Considerations in Development of Tantalum Base Alloys," from Refractory Metal Alloys Metallurgy and Technology, Editors J. Machlin, R. T. Begley, and E. D. Weisert, Plenum Press, New York, 1968.

4) R. W. Buckman, Jr., "Development of Advanced High Strength Tantalum Base Alloys" Part I - Screening Investigation - WANL-PR-71-001, NASA CR-120818.

5) R. W. Buckman, Jr. and J. J. Hetherington, "Apparatus for Determining Creep Behavior Under Conditions of Ultra High Vacuum," Review of Scientific Instruments, Vol. 37, No. 8, pp. 999-1003, August 1966.